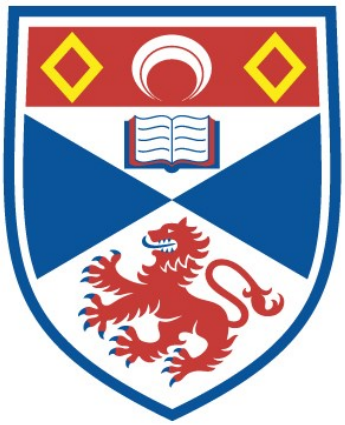
STRUCTURAL STUDIES OF SOME HETEROCYCLIC AND ORGANOMETALLIC COMPOUNDS

Hazel Diane Holden

A Thesis Submitted for the Degree of PhD
at the
University of St Andrews



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Structural Studies of Some Heterocyclic and Organometallic Compounds.

being a thesis

presented by

Hazel Diane Holden, B.Sc.

to the

University of St. Andrews

in application for

the degree of Doctor of Philosophy.

St. Andrews September 1981

UNIVERSITY OF ST. ANDREWS

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Abstract.

Single crystal X-ray work was carried out for four heterocyclic compounds: prior to this, definitive structures could not be written for any of these. The heterocyclic compounds reported are: 5,6-dihydro-1-thiobenzoylmethylene-1H-thiazolo[2,3-c][1,2,4] thiadiazole, (I); 4-N-(4′-bromophenylamino)-3-butylfuroxan, (II); 4,4′-methylenebis(1,3,5-trimethyl-4-imidazolin-2-one), (III), and 4-phenyl-3-phenylamino -1,2,4-thiadiazoline-5-one, (IV).

These are then discussed, along with work which has been carried out for similar compounds. Trithiapentalenes and analogues having linear multisulphur systems are mentioned as criteria for deciding whether the NSS skeleton in (I) is best considered as N-S...S or N-S-S.

Following this, the X-ray structure determinations of bis(4-methylpyridine)hydrogen(I) tetraphenylborate, (V), and bis(triphenylarsineoxide)hydrogen(I) tetrafluoroborate, (VI), are reported. Both (V) and (VI) are bis-cations which have strong hydrogen bonding. Other compounds with short O...H...O and N...H...N distances are mentioned, stabilities of such bis-cations compared to those of the monocations, and $(\text{amine-halogen-amine})^{+}$ complexes discussed.

The final two X-ray structure determinations are for the compounds 1,1,1,3,3,3-hexaphenyldisilazane and μ-nitrido-bis (triphenylphosphorus)((I) thiocyanate.

MNDO was used to calculate force constants for bending of linear molecular pseudohalides. The aim of such work was to rationalise 'unusual' geometries in some silicon-containing compounds via Second Order Jahn-Teller distortions, postulating that such compounds are easily distorted along a 'soft' normal vibration coordinate.

Declaration.

I declare that this thesis is a record of my own experiments, that it is my own composition, and that it has not previously been presented in application for a higher degree.

The work was carried out in the Department of Chemistry of the University of St. Andrews under the direction of Dr. C. Glidewell.

Certificate.

I hereby certify that Miss Hazel Diane Holden, B.Sc., has spent eleven terms at research work under my supervision, has fulfilled the conditions of the resolution of the University Court 1967, No. 1, and is qualified to submit the accompanying thesis in application for the degree of Doctor of Philosophy.

Research Supervisor

Acknowledgements.

I would like to express thanks to my parents for helping me to begin my university career and for their unfailing encouragement since then; to the University Court of the University of St. Andrews for their generosity in awarding me the 'Sir James Irvine Memorial Scholarship' which has financed my post-graduate work; and to Professor P.A.H. Wyatt, the Irvine Professor of Chemistry for the excellent laboratory and library facilities available here at St. Andrews.

The technical staff, laboratory stewards and department librarian, Mrs. S. Johnson have always been obliging. Jim Bews was responsible for the running of the MNDO programme system, and David Liles for the various crystallographic programmes which were used. Members of the Computing Laboratory, in particular Dr. Ian Begg, have always been most helpful.

Alastair Cuthbertson, David Liles, Mr. J.S. McKechnie and Peter Pogorzelec were always ready to enter into stimulating discussions.

For his never-ending enthusiasm and encouragement, the final, but greatest word of appreciation should go to Dr. Christopher Glidewell. Without him, this work would not have been possible, and I thank him very much indeed.

Abstract.

Single crystal X-ray work was carried out for four heterocyclic compounds: prior to this, definitive structures could not be written for any of these. The heterocyclic compounds reported are: 5,6-dihydro-1-thiobenzoylmethylene-1H-thiazolo[2,3-c][1,2,4] thiadiazole, (I); 4-N-(4'-bromophenylamino)-3-butylfuroxan, (II); 4,4'-methylenebis(1,3,5-trimethyl-4-imidazolin-2-one), (III), and 4-phenyl-3-phenylamino -1,2,4-thiadiazoline-5-one, (IV).

These are then discussed, along with work which has been carried out for similar compounds. Trithiapentalenes and analogues having linear multisulphur systems are mentioned as criteria for deciding whether the NSS skeleton in (I) is best considered as N-S...S or N-S-S.

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Contents.

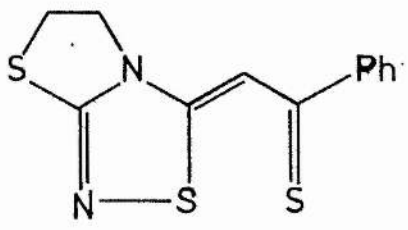
Structural Studies of Some Heterocyclic and Organometallic Compounds.

Chapter One.

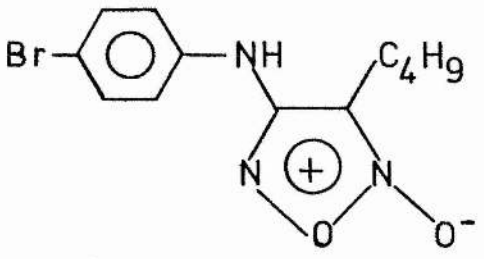
An Introduction.

The advent of more sophisticated X-ray diffractometers, and large computers, has greatly increased the number of single-crystal X-ray structure determinations being added to the chemical literature each year. Eight such structural determinations form the focal points for the work here presented.

This thesis may be considered in three major sections, the first of which is concerned with X-ray structure determinations for four organic compounds, (I), (II), (III) and (IV). Full structural investigations of these were undertaken simply because, prior to this X-ray work, definitive structures could not be written for any of the compounds.



(I)



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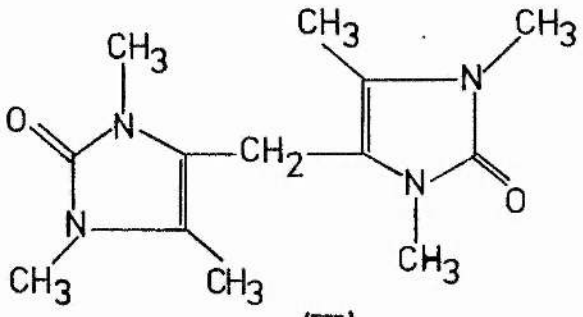
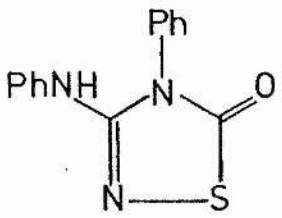
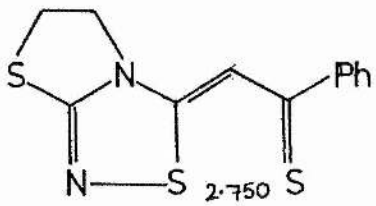


(III)

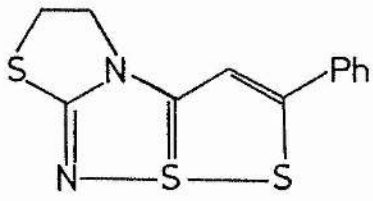


(IV)

5,6-Dihydro-1-thiobenzoylmethylene-1H-thiazolo[2,3-c][1,2,4] thiadiazole, (I), was of interest because it was not known whether the best representation of the structure of this compound was indeed (I), or (I(ii)). An S....S distance of 2.750 Å confirmed (I).

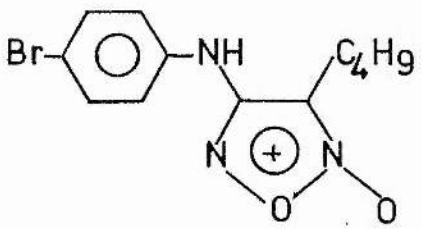


(I)

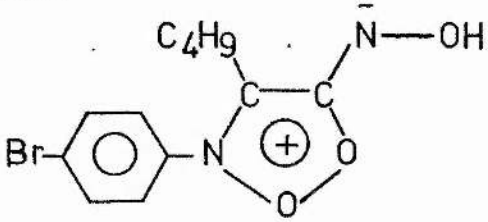


(I(ii))

Before single crystals of (II) were examined, this compound's solid-state structure was unknown. Woodcock[1] had suggested it was a sydnone oxime, (II(ii)), but the subsequent structural investigation reported in this thesis refuted this.



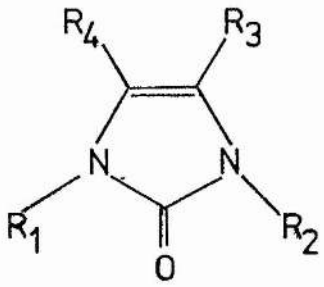
(II)



(II(ii))

Compound (III) was the very unexpected product of the reaction of 1,3-dimethylurea with butan-2,3-dione, carried out by Drs. A.R. Butler and I. Hussein. The usual products of reaction of acyloins with urea under similar conditions are 4-imidazolin-2-ones, (V)[2].

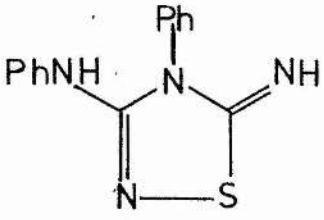
(V)



The stoichiometry of (III), $C_{13}H_{20}N_4O_2$ and the extremely simple ^1H and ^{13}C n.m.r. spectra which indicated two-fold molecular symmetry in solution, showed that (III) is an unusual product. Without X-ray work, the structure of (III) would probably have remained an enigma.

The fourth compound (IV) is generally known as 'Dost's Keto Compound'[3]. It is made from 'Hector's Base'[4,5] (VI), and was of interest to us as part of a series of work on derivatives and adducts of Hector's Base.

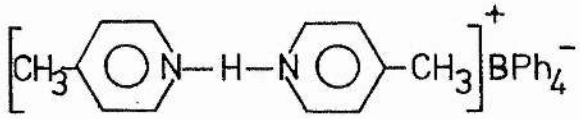
(VI)



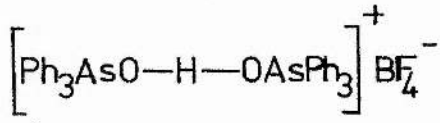
These four crystal structure determinations are given in Chapter Two; the following chapter is a discussion of these and some related heterocyclic compounds.

The next section follows the same format of a report of the X-ray work done, and then a discussion of the compounds, namely why they were of interest and any similar compounds which have been reported.

Chapter Four is concerned with structures of two compounds which contain bis-cations: these are bis(4-methylpyridine)hydrogen(I) tetraphenylborate (VII), and bis(triphenylarsineoxide)hydrogen(I) tetrafluoroborate, (VIII).



(VII)



(VIII)

Characterisation of (VII) was carried out, to show whether this bis-cation did indeed exist in the solid-state. Its presence had been suspected by Glidewell and Shepherd[6], but not proven. It was still possible that in the solid the simple 4-methylpyridinium cation was present, together with the neutral amine as solvent of crystallisation. The X-ray work described in this thesis served to confirm the existence of the bis-cation.

In Chapter Five, this cation is compared to other cationic complexes which neutral amines form with the halogens. Planarity or non-planarity of such complexes is then explored, using MNDO[7] (Modified Neglect of Diatomic Overlap) calculations. For $(4\text{-mepy})_2H^+$ and $(4\text{-mepy})_2Cl^+$ (4-mepy = 4-methylpyridine)energy barriers to planarity are calculated to be so small, that lattice forces will suffice to make the complexes planar. Steric repulsions between ortho hydrogens are the most probable reason for the slight preference for a non-planar twisted cation, when the calculations are carried out for isolated cations in the gas phase.

Stability of ions in the solid is also discussed in Chapter Five via a Born-Haber cycle: the deductions made are that inherently unstable small cations and anions may be stabilised by a small counter ion, whilst for an inherently unstable large ion, a large counter ion will be most effective in rendering stability to its partner.

The bis(triphenylarsineoxide)hydrogen(I) cation has been previously reported[8]. Both of these bis-cations have very strong hydrogen bonds. Chapter Five ends by comparing O...H...O and N...H...N distances in other strongly hydrogen bonded species, and with some comments on hydrogen bonding in solids.

The third and final section comprises three chapters, in the first of which two crystal structure determinations are described. These are for two compounds which have the general formula R_3XYZR_3: 1,1,1,3,3,3-hexaphenyldisilazane (IX), and compound (X) which is μ-nitrido-bis(triphenylphosphorus)(I) thiocyanate.

$Ph_3SiNHSiPh_3$ (IX)

$[Ph_3PNPPh_3]^+SCN^-$ (X)

The central chapter of this section describes MNDO calculations carried out in an attempt to find force constants for bending of linear molecular pseudohalides. A description of the 'Second Order Jahn-Teller' (SOJT) effect and its effectiveness in rationalising many molecular geometries completes the trilogy. Also under discussion in Chapter Eight are geometries of similar R_3XYZR_3 compounds which are reported in the literature, and how Chapter Seven's MNDO work is in accord with experimentally determined geometries of some pseudohalide molecules. Of particular interest has been the concept of isoelectronic XYZ fragments such as Si-O-Si, $P-N-P^+$ and P=C=P having low energy barriers to linearity, which allows a wide range of possible geometries in the solid state.

For clarity, I have reported each crystal structure as a separate entity. This means that no cross references are necessary. Following the report of each crystal structure are drawings of the molecules, the packing arrangements in the unit cells and tables of fractional atomic coordinates and geometries.

Chapter One.

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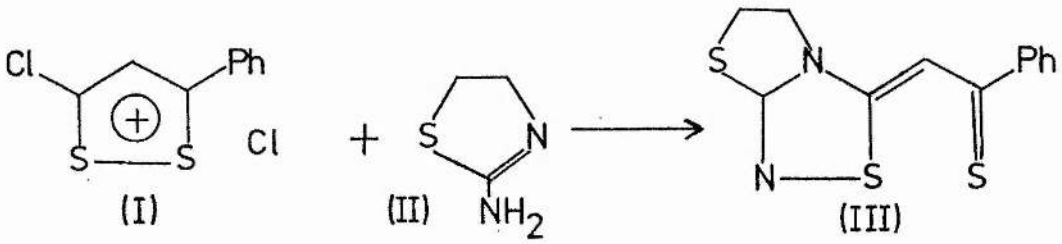
Chapter Two.

Crystallographic Structure Determinations of Some Organic Heterocyclic Molecules.

I. 5,6-Dihydro-1-thiobenzoylmethylene-1H-thiazolo[2,3-c][1,2,4]thiadiazole.

Preparation.

Crystals of this compound were kindly provided by Professor D.H. Reid and Dr.J. Mitchell. These were prepared by the reaction of 3-chloro-5-phenyl-1,2-dithiol-1-ium chloride (I) with 2-amino-4,5-dihydrothiazole (II)[1,2]:



Crystal Data.

$C_{12}H_{10}N_2S_3$ (III) is monoclinic, space-group C2/c (C_{2h}^6, No. 15). a = 23.316(11), b = 8.187(11) c = 13.127(10) Å, β = 103.73(5)°, U = 2434.28 Å^3, Mr=278.40, Dc=1.519 g.cm^{-3}. for Z = 8. F(000) = 1152.00. Mo-K_α radiation used with λ = 0.71069 Å, μ(Mo-K_α) = 5.21 cm^{-1}.

Data Collection and Structure Solution.

The intensities of 1426 unique reflections in the quadrant +h, +k, ±l, with $2^{\circ} \leqslant \theta \leqslant 30^{\circ}$, k = 0 - 10 ($0.000^{\circ} \leqslant \mu \leqslant 25.723^{\circ}$), were measured, using a Stoe STADI-2 diffractometer in the $\omega - 2\theta$ scan mode, and graphite monochromatised Mo-K_{α} radiation; the scan speed was $0.0167^{\circ}s^{-1}$. in ω, and backgrounds were measured at each end of the peak scan for a time of (scan time/2). Standard reflections, measured every fifty reflections, showed only small random deviations from their means. Lorentz and polarisation corrections were then applied to the data, but no corrections for absorption made.

Systematic absences were: in hkl, h + k ≠ 2n; in h0l, l ≠ 2n and in 0 k 0, k ≠ 2n, thus indicating space-group Cc or C2/c. Structure solution and refinement confirmed C2/c.

The structure was solved in C2/c by direct methods, using the SHELX 76 programme system[3]. The best E map gave positions for all non-hydrogen atoms, and three cycles of full-matrix least-squares refinement gave an $R(= \Sigma \Delta / \Sigma F_o, \quad \Delta = |F_o - F_c|)$ of 0.2357 and an Rg $[= (\Sigma \omega \Delta^2 / \Sigma \omega F_o^2)^{1/2}]$ of 0.2625.

Introduction of anisotropic thermal parameters for the S and N atoms reduced R to 0.1269 and Rg to 0.1527, and for the non-phenyl C atoms, R to 0.1196 and Rg to 0.1453. The unique H(5) atom and the phenyl H atoms were located from the difference map. Introducing these atoms with a common isotropic thermal parameter for phenyl

hydrogens and anisotropic thermal parameters for all non-hydrogen atoms, reduced R to 0.0900 and Rg to 0.0958 for 1162 intensity data having $F_o \geqslant 6\sigma(F_o)$. The four methylene hydrogen atoms attached to C(3) and C(4) were not unambiguously identified in the difference map, and so were placed in calculated positions with d(C-H) = 1.080 Å and angle H-C-H = 109.5°. All four were tied to a common isotropic thermal parameter, and in this way R was brought to a final value of 0.0880 and Rg to 0.0916. Releasing the positional constraints on the methylene hydrogens did bring Rg to 0.0910, but the geometrical parameters were then unacceptable, hence their positions are not well determined by the X-ray data.

In the final cycles of refinement, 175 parameters were varied simultaneously, that is, 69 positional coordinates, 102 anisotropic thermal parameters, three isotropic thermal parameters and one overall scale factor. Complex neutral-atom scattering factors were used for all atoms[4,5,6] and the reduction in Rg was significant at the 99.5 % level[7] for each stage of refinement, except removal of geometrical constraints for the H atoms on C(3) and C(4).

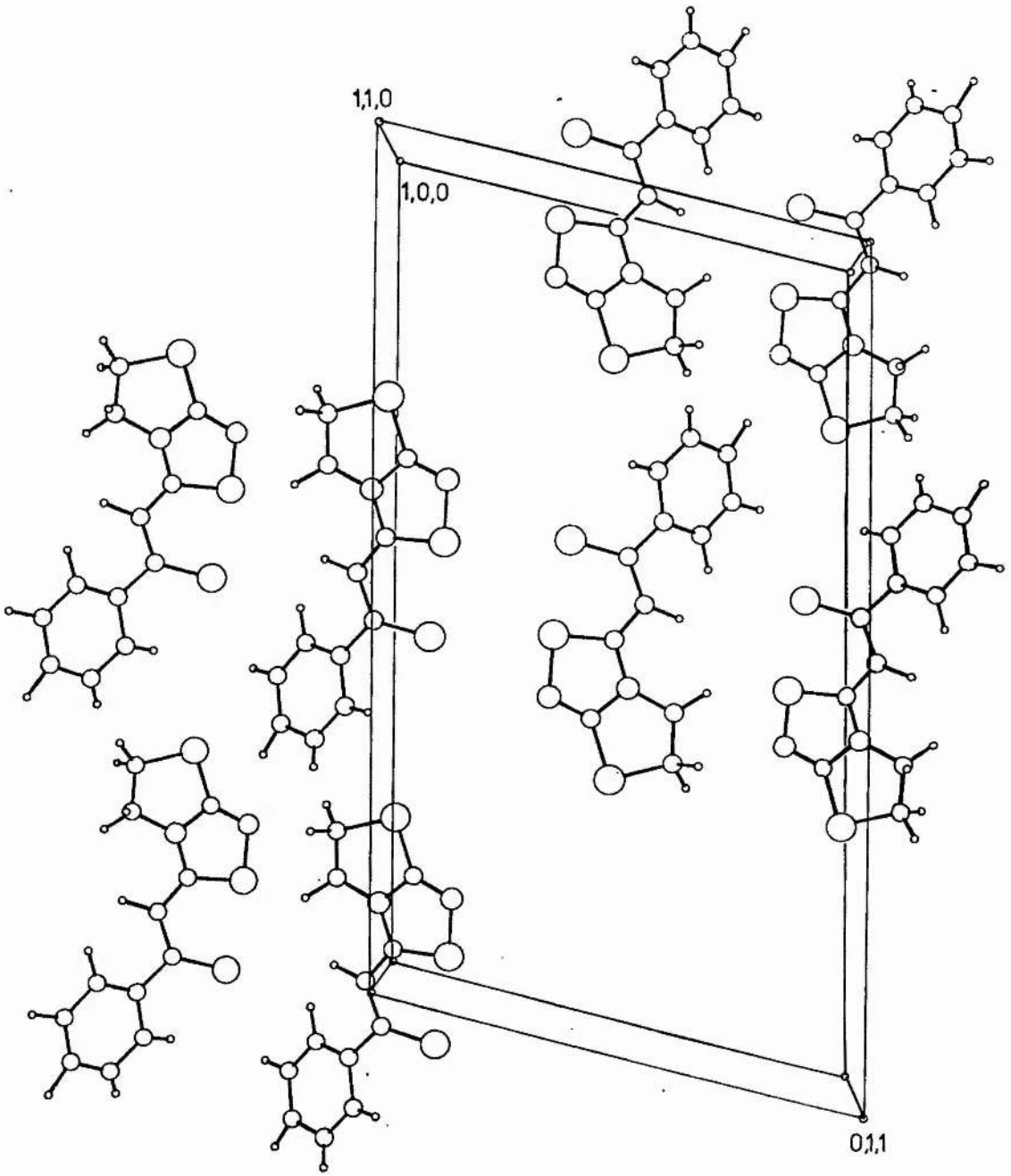


Figure 2.1.2. The contents of the unit cell. The view is down b.

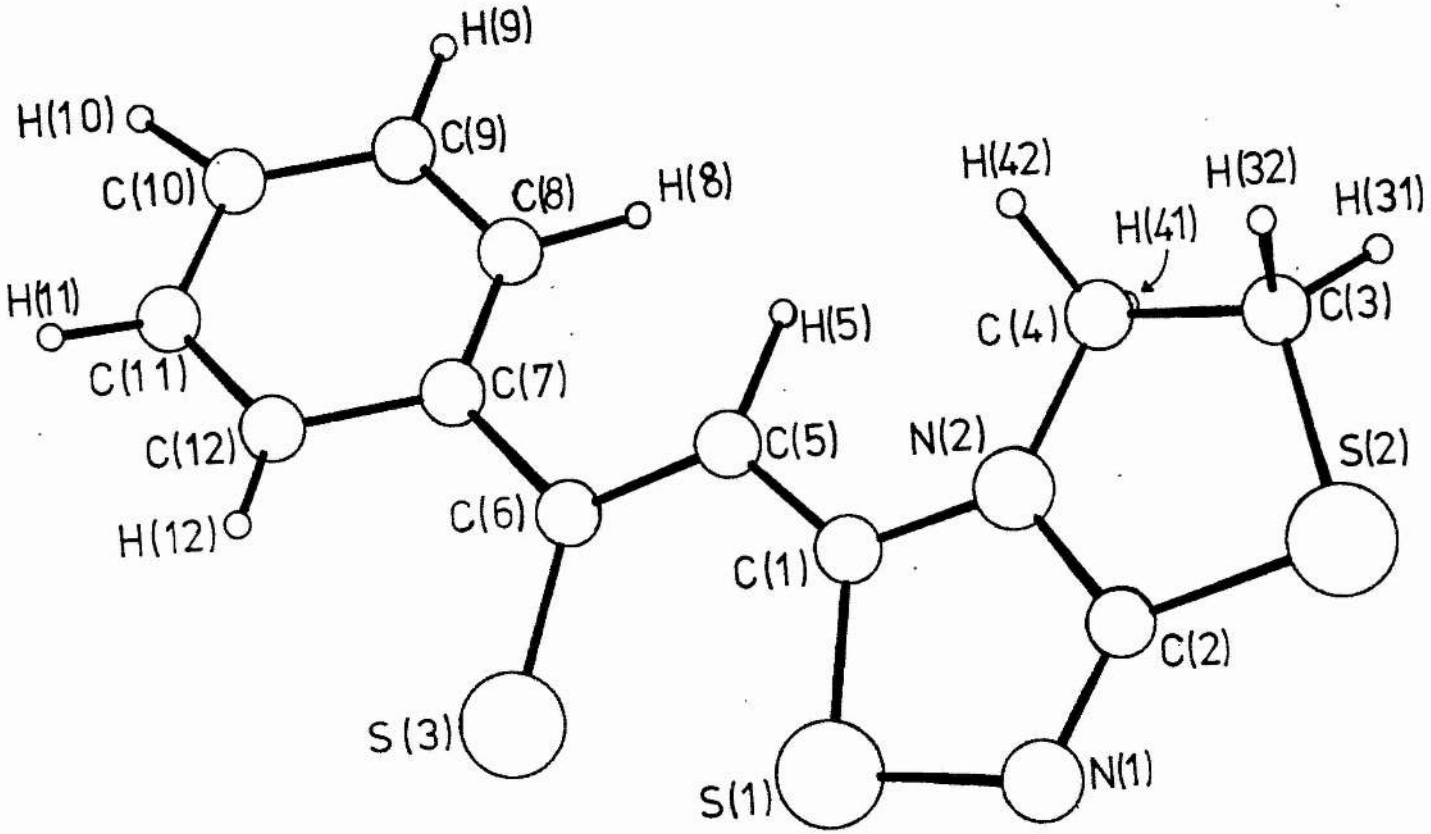


Figure 2.1.1. The molecule, with the numbering scheme.

Table 2.1.1. Fractional Atomic Coordinates (x 10^4; for H x 10^3).

Atom	x	y	z
S(1)	9618(1)	3616(4)	3643(2)
S(2)	8099(1)	3757(5)	4815(2)
S(3)	773(2)	2610(5)	3966(2)
N(1)	8895(4)	4167(13)	3572(7)
N(2)	9210(4)	2968(11)	5184(6)
C(1)	9730(5)	2806(13)	4887(8)
C(2)	8757(5)	3652(17)	4422(8)
C(3)	8461(6)	3066(21)	6115(12)
C(4)	9055(5)	2448(18)	6137(8)
C(5)	234(5)	2117(15)	5512(8)
C(6)	751(5)	1964(14)	5163(8)
C(7)	1278(4)	1219(14)	5902(8)
C(8)	1239(5)	181(15)	6698(8)
C(9)	1735(5)	-487(15)	7354(9)
C(10)	2289(5)	-56(17)	7240(10)
C(11)	2350(5)	974(17)	6451(10)
C(12)	1851(5)	1667(16)	5770(10)
H(31)	820(1)	216(2)	639(1)
H(32)	852(1)	411(2)	663(1)
H(41)	903(1)	113(2)	615(1)
H(42)	938(1)	287(2)	681(1)
H(5)	17(7)	132(22)	624(13)
H(8)	84(5)	-26(13)	686(8)
H(9)	174(4)	-155(14)	796(8)
H(10)	273(5)	-67(13)	766(8)
H(11)	266(5)	188(14)	641(8)

H(12)	184(5)	254(14)	524(8)

Table 2.1.2. Geometry of the Molecule.

(a) Distances(Å).

S(1) ... S(3)	2.750(5)				
S(1) - N(1)	1.727(10)	C(8) - C(9)	1.379(15)		
S(1) - C(1)	1.723(11)	C(9) - C(10)	1.382(17)		
S(2) - C(2)	1.734(11)	C(10) - C(11)	1.369(17)		
S(2) - C(3)	1.806(14)	C(11) - C(12)	1.409(17)		
S(3) - C(6)	1.670(11)	C(3) - H(31)	1.080(0)		
N(1) - C(2)	1.302(13)	C(3) - H(32)	1.080(0)		
N(2) - C(1)	1.365(12)	C(4) - H(41)	1.080(0)		
N(2) - C(2)	1.388(13)	C(4) - H(42)	1.080(0)		
N(2) - C(4)	1.447(12)	C(5) - H(5)	1.197(166)		
C(1) - C(5)	1.384(14)	C(8) - H(8)	1.061(108)		
C(3) - C(4)	1.467(15)	C(9) - H(9)	1.180(110)		
C(5) - C(6)	1.393(14)	C(10) - H(10)	1.165(108)		
C(6) - C(7)	1.503(14)	C(11) - H(11)	1.051(112)		
C(7) - C(8)	1.367(15)	C(12) - H(12)	0.992(112)		
C(7) - C(12)	1.433(15)				

(b) Angles(°).

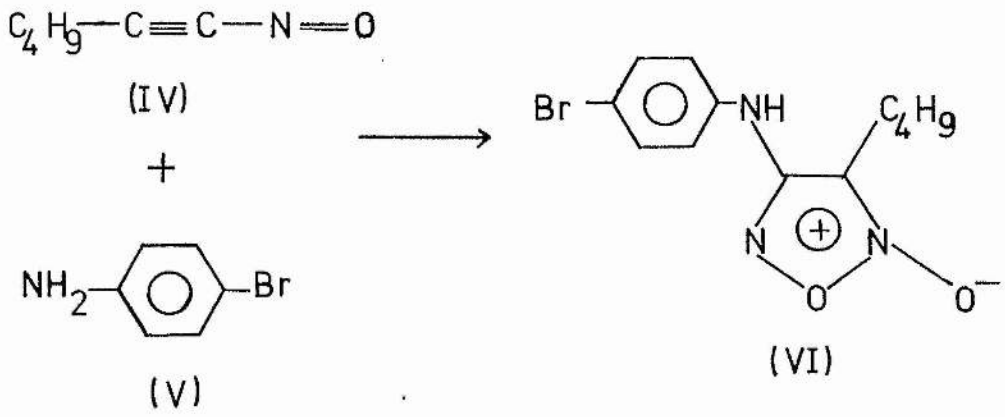
N(1) - S(1) - C(1)	94.5(5)	S(3) - C(6) - C(7)	122.1(8)
C(2) - S(2) - C(3)	91.0(5)	C(5) - C(6) - C(7)	117.7(9)
S(1) - N(1) - C(2)	107.7(8)	C(6) - C(7) - C(8)	123.5(10)
C(1) - N(2) - C(2)	114.2(8)	C(6) - C(7) - C(12)	117.5(10)
C(2) - N(2) - C(4)	115.9(9)	C(8) - C(7) - C(12)	119.0(10)

S(1) - C(1) - N(2)	106.7(7)	C(7) - C(8) - C(9)	121.7(11)
S(1) - C(1) - C(5)	129.5(8)	C(7) - C(8) - H(8)	125.6(61)
N(2) - C(1) - C(5)	123.8(9)	C(9) - C(8) - H(8)	112.5(61)
S(2) - C(2) - N(2)	112.4(8)	C(8) - C(9) - C(10)	119.8(12)
N(1) - C(2) - N(2)	116.6(9)	C(8) - C(9) - H(9)	126.0(52)
S(2) - C(3) - C(4)	110.7(9)	C(10) - C(9) - H(9)	113.7(51)
S(2) - C(3) - H(31)	111.3(5)	C(9) - C(10) - C(11)	120.5(12)
S(2) - C(3) - H(32)	107.9(6)	C(9) - C(10) - H(10)	125.6(54)
N(2) - C(4) - C(3)	108.1(10)	C(11) - C(10) - H(10)	113.0(52)
N(2) - C(4) - H(41)	109.5(7)	C(10) - C(11) - C(12)	120.6(12)
N(2) - C(4) - H(42)	109.6(6)	C(10) - C(11) - H(11)	132.1(61)
C(1) - C(5) - C(6)	121.4(10)	C(12) - C(11) - H(11)	99.4(60)
C(1) - C(5) - H(5)	116.7(77)	C(7) - C(12) - C(11)	118.3(11)
C(6) - C(5) - H(5)	119.6(79)	C(7) - C(12) - H(12)	113.2(66)
S(3) - C(6) - C(5)	120.2(9)	C(11) - C(12) - H(12)	128.1(66)

II. 4-N-(4'-bromophenylamino)-3-butylfuroxan.

Preparation.

Crystals of the heterocyclic base, $C_{12}H_{14}N_3O_2Br$ (VI) suitable for X-ray diffraction work were kindly provided by Dr. D.J. Woodcock. These were prepared by reaction of 1-nitrosohex-1-yne (IV) with 4-bromoaniline (V): the crystals are colourless, M.Pt. 153-154°C.(8a,8b)



Crystal Data.

$C_{12}H_{14}N_3O_2Br$ (VI) is monoclinic, space-group C2/c (C_{2h}^6, No. 15). a = 24.154(3), b = 12.362(2), c = 9.578(2) Å, β = 106.95(1)°, U = 2735.6 Å^3, Mr = 312.17, Dc = 1.516 g.cm^{-3}. for Z = 8. F(000) = 1264.00. Mo-Kα radiation used with λ= 0.71069 Å, μ(Mo-Kα) = 29.19 cm^{-1}.(9a,9b)

Data Collection and Structure Solution.

Intensity data for 3239 reflections in the hemisphere $\pm h$, $\pm k$, $+l$, with $3^{o} \leqslant \theta \leqslant 27^{o}$ were collected in the ω-2θ scan mode on a Philips PW 1100 diffractometer using graphite monochromatised Mo-K_{α} radiation and a crystal of dimensions 0.38 x 0.40 x 0.45 mm. After merging the data in Cc, 1817 unique data remained with $F_o \geqslant 6\sigma(F_o)$. X-ray fluorescence was thought to be the cause of rather high background readings.

Systematic absences were: in hkl, $h + k \neq 2n$; h0l, $l \neq 2n$, indicating space-group Cc or C2/c. The latter was confirmed by structure solution and the ensuing satisfactory refinement.

Using the SHELX 76 programme system[3] a Patterson synthesis showed only one type of bromine atom to be present and calculated the coordinates. A difference map then calculated positions for all non-hydrogen atoms and for the amino and aryl hydrogens. Full-matrix least-squares refinement successively introducing anisotropic thermal parameters initially for bromine, then other non-hydrogen atoms, followed by an $\omega = [\sigma^2(F)]^{-1}$ weighting scheme reduced R to 0.0930 and Rg to 0.0806.

The geometry of the heterocyclic ring is well defined, but there were four plausible atom permutations of this heterocycle, assuming that the carbon chain of the 1-nitrosohex-1-yne has remained unbroken throughout reaction. The R factors for these isomers were compared

(see Table 2.2.1) indicating (VI) to be the correct structure.

Figure 2.2.1. Possible Isomers.

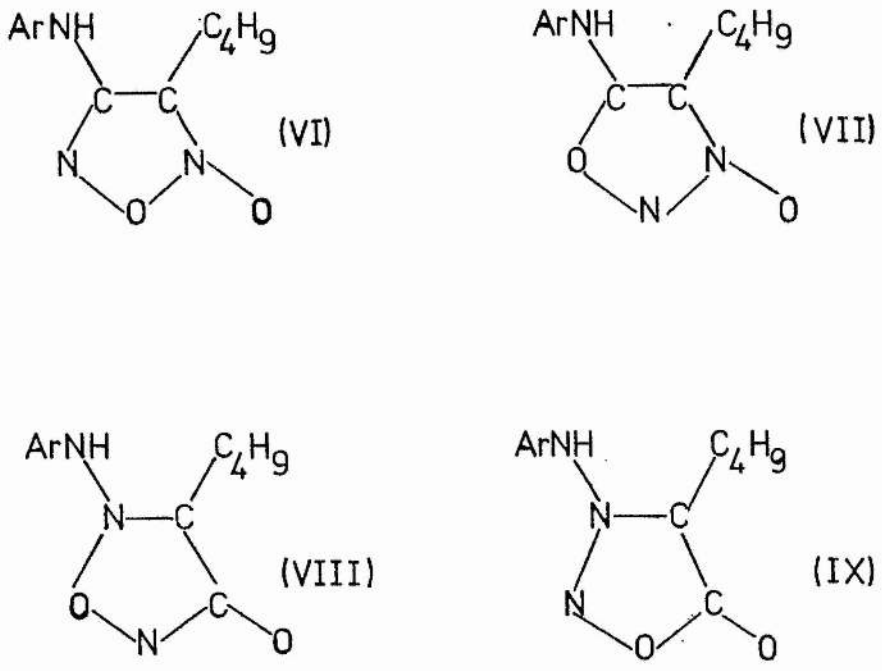


Table 2.2.1. Isomer Identification.

Isomer	VI	VII	VIII	IX
R	0.0930	0.0978	0.1042	0.0992
Rg	0.0805	0.0854	0.0918	0.0860

By this stage of the refinement, the difference map showed the two central carbon atoms of the butyl group to be disordered. The end carbons of the butyl chain were given isotropic thermal parameters and the four partial atoms were given a common isotropic thermal

parameter. When constrained to sum to unity the site occupation factors of the two conformations refined to 0.81(1) and 0.19(1). The final values of R, the weighted index Rw ($=\sum\Delta\omega^{1/2}/\sum F_o\omega^{1/2}$) and Rg were 0.0947, 0.0797 and 0.0828 respectively for 165 parameters varying simultaneously. These comprised 75 positional parameters, 84 anisotropic thermal parameters, 4 isotropic thermal parameters, one site occupation factor and one overall scale factor. Complex neutral-atom scattering factors were used throughout[4,5,6].

Figure 2.2.3. The unit cell, viewed down b. The furoxan molecules are almost planar.

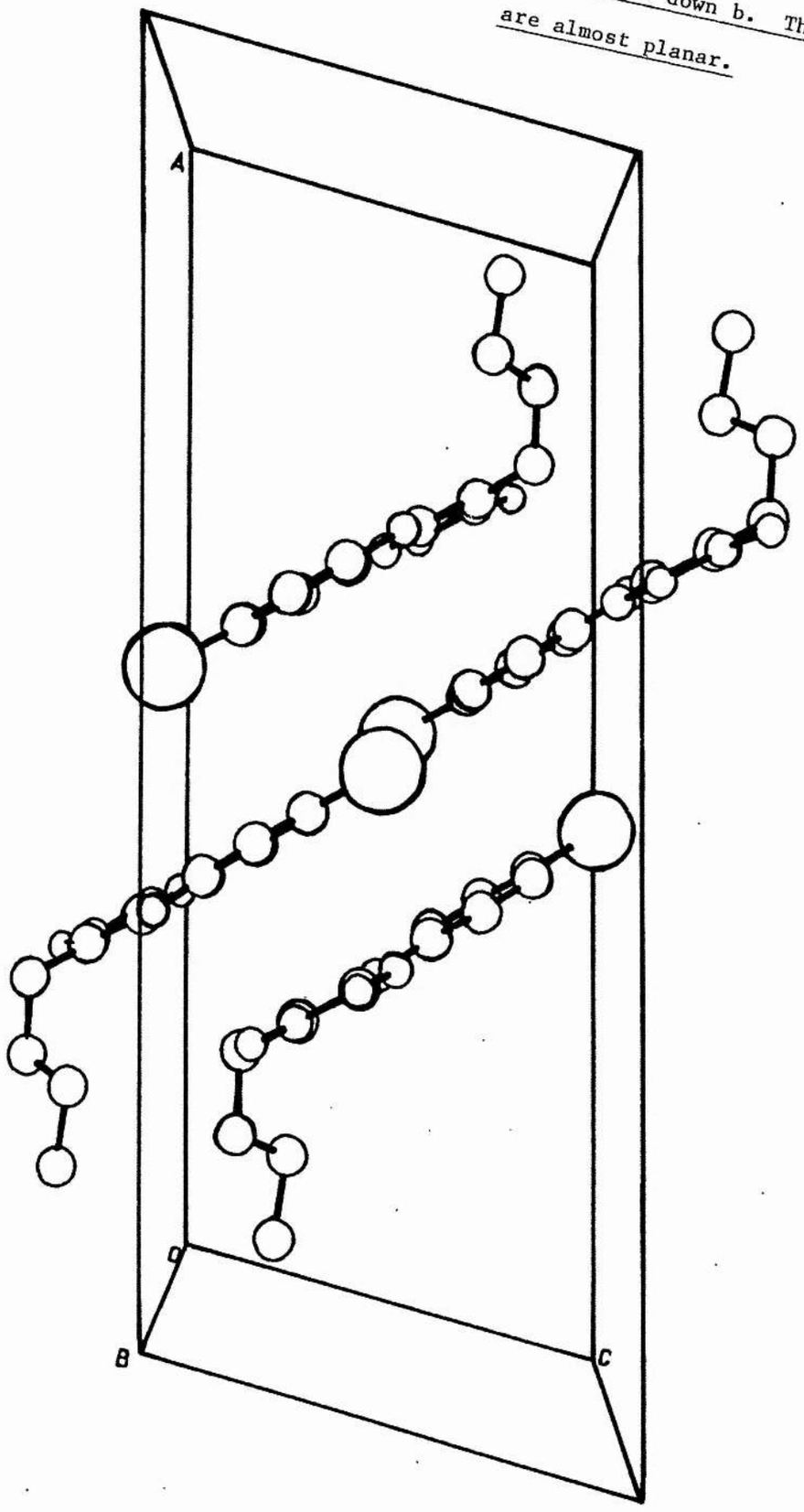


Figure 2.2.2. The furoxan molecule, showing the disordered butyl group.

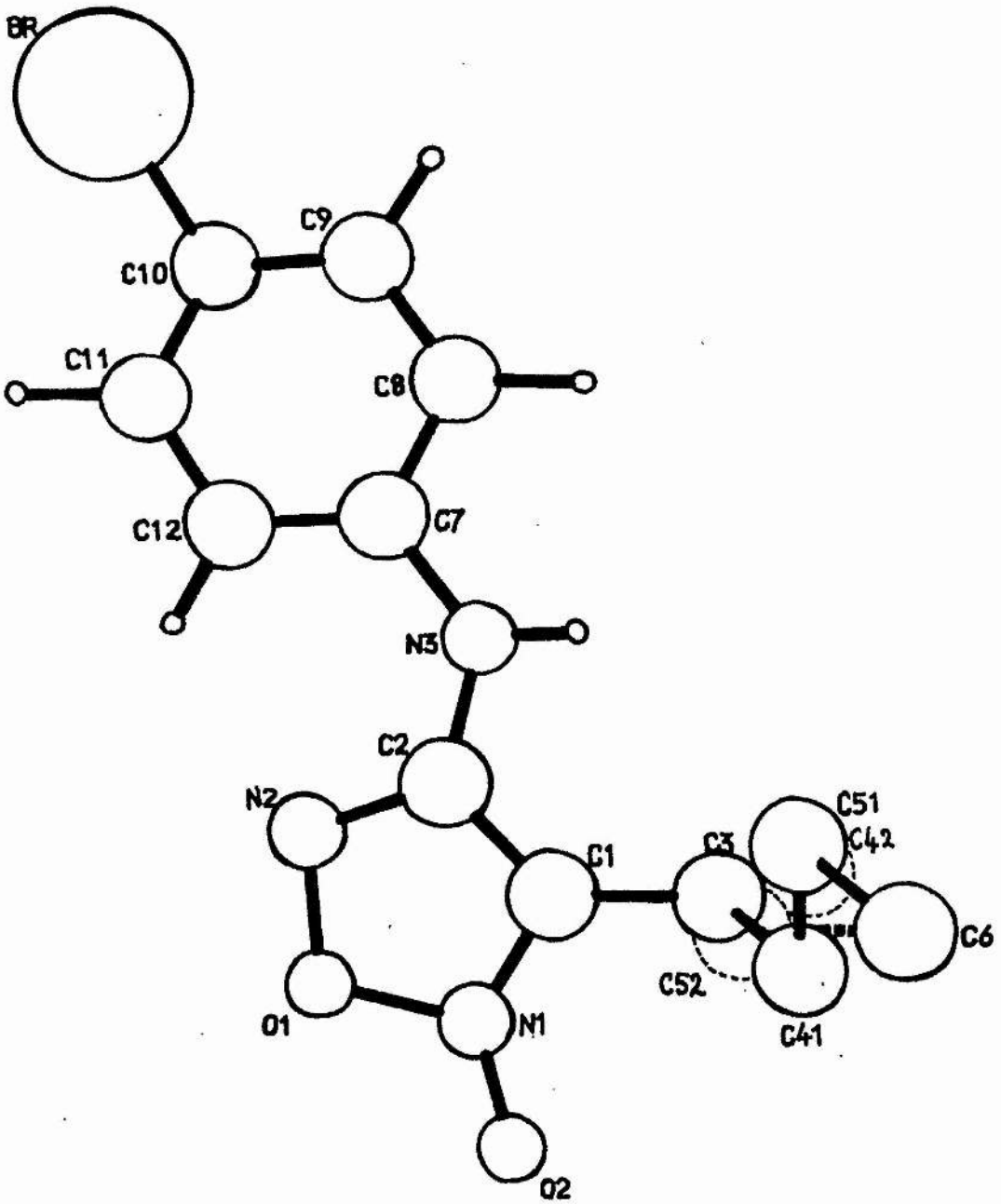


Table 2.2.2. Fractional Atomic Coordinates(x 10^4; for H x 10^3).

Atom	x	y	z
Br	4837(0)	2808(1)	9826(1)
O(1)	3082(2)	7826(4)	4335(6)
O(2)	2454(3)	8222(4)	2124(7)
N(1)	2669(3)	7516(5)	3002(7)
N(2)	3292(2)	6894(4)	5130(7)
N(3)	3129(3)	5031(5)	4693(7)
C(1)	2634(3)	6438(5)	3006(7)
C(2)	3024(3)	6098(5)	4327(7)
C(3)	2237(3)	5862(6)	1743(8)
C(41)	1565(4)	6029(11)	1640(12)
C(42)	1686(14)	5412(36)	2000(50)
C(51)	1456(5)	5283(10)	2712(14)
C(52)	1378(11)	5973(38)	2981(42)
C(6)	755(4)	5462(10)	2428(13)
C(7)	3548(3)	4583(6)	5905(8)
C(8)	3580(3)	3445(6)	5944(9)
C(9)	3971(4)	2946(7)	7122(10)
C(10)	4327(3)	3546(7)	8220(8)
C(11)	4295(3)	4647(8)	8198(10)
C(12)	3907(3)	5177(7)	7025(9)
H(10)	290(3)	465(6)	400(10)
H(11)	331(4)	299(7)	490(10)
H(12)	394(4)	208(7)	709(11)
H(13)	464(3)	485(7)	796(11)
H(14)	396(3)	602(8)	694(9)

Table 2.2.3. Geometry of the Furoxan compound.

(a) Bond Distances(Å).

O(1)	- N(1)	1.424(7)	N(3)	- C(7)	1.413(8)
O(2)	- N(1)	1.219(7)	C(7)	- C(8)	1.409(9)
N(1)	- C(1)	1.355(8)	C(8)	- C(9)	1.387(10)
C(1)	- C(2)	1.404(8)	C(9)	- C(10)	1.367(10)
N(2)	- C(2)	1.299(7)	C(10)	- C(11)	1.364(11)
O(1)	- N(2)	1.392(6)	C(11)	- C(12)	1.399(10)
C(1)	- C(3)	1.488(9)	C(12)	- C(7)	1.378(9)
C(3)	- C(41)	1.612(10)	N(3)	- H(10)	0.86(7)
C(3)	- C(42)	1.527(18)	C(8)	- H(11)	1.16(9)
C(41)	- C(51)	1.460(13)	C(9)	- H(12)	1.08(9)
C(42)	- C(52)	1.523(21)	C(11)	- H(13)	0.96(8)
C(51)	- C(6)	1.649(12)	C(12)	- H(14)	1.06(9)
C(52)	- C(6)	1.575(19)	Br	- C(10)	1.899(7)
N(3)	- C(2)	1.369(8)			

(b) Interbond Angles(°).

O(1)	- N(1)	- O(2)	118.2(5)	C(7)	- C(8)	- C(9)	118.9(8)
O(1)	- N(1)	- C(1)	107.1(5)	C(8)	- C(9)	- C(10)	120.8(7)
O(2)	- N(1)	- C(1)	134.6(6)	C(9)	- C(10)	- C(11)	120.6(7)
N(1)	- C(1)	- C(2)	105.9(6)	C(10)	- C(11)	- C(12)	120.1(8)
N(1)	- C(1)	- C(3)	120.1(6)	C(11)	- C(12)	- C(7)	119.9(8)
C(2)	- C(1)	- C(3)	133.9(6)	C(12)	- C(7)	- C(8)	119.7(7)
N(2)	- C(2)	- C(1)	113.2(6)	N(3)	- C(7)	- C(12)	124.6(7)
N(3)	- C(2)	- C(1)	123.0(6)	C(2)	- N(3)	- H(10)	107(5)

O(1)	- N(2)	- C(2)	105.3(5)	C(7)	- N(3)	- H(10)	124(5)
N(2)	- C(2)	- N(3)	123.7(6)	C(7)	- C(8)	- H(11)	117(4)
N(1)	- O(1)	- N(2)	108.4(5)	C(9)	- C(8)	- H(11)	124(4)
C(1)	- C(3)	- C(41)	112.8(7)	C(8)	- C(9)	- C(12)	113(5)
C(1)	- C(3)	- C(42)	115.3(18)	C(10)	- C(9)	- H(12)	126(5)
C(3)	- C(41)	- C(51)	105.1(9)	C(10)	- C(11)	- H(13)	102(6)
C(3)	- C(42)	- C(52)	122.6(27)	C(12)	- C(11)	- H(13)	97(6)
C(41)	- C(51)	- C(6)	100.7(9)	C(11)	- C(12)	- H(14)	117(5)
C(42)	- C(52)	- C(6)	101.6(18)	C(7)	- C(12)	- H(14)	122(5)
C(2)	- N(3)	- C(7)	128.5(6)	Br	- C(10)	- C(9)	118.5(6)
N(3)	- C(7)	- C(8)	115.7(7)	Br	- C(10)	- C(11)	120.8(6)

III. 4,4´-Methylenebis(1,3,5-trimethyl-4-imidazoline-2-one).

Preparation.

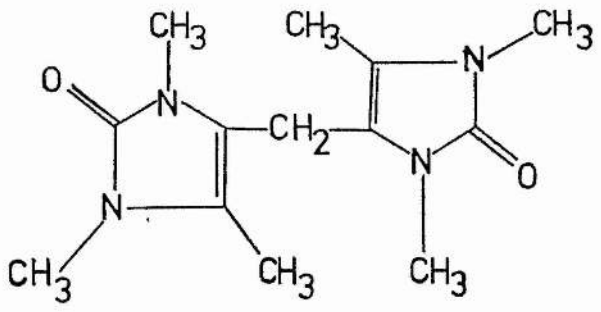
1,3-Dimethylurea reacts with butan-2,3-dione in refluxing benzene containing about 10 % CF_3COOH to give a compound of stoichiometry $C_{13}H_{20}N_4O_2$.

$$2\ CH_3NH.CO.NHCH_3 + 2\ CH_3.CO.CO.CH_3 \xrightarrow[\text{in benzene.}]{10\%\ CF_3COOH} C_{13}H_{20}N_4O_2$$

(X)

Although the very simple proton-coupled ^{13}C n.m.r. spectrum indicates a two-fold element of symmetry within the molecule, it is not possible to suggest an unequivocal structure on the basis of spectroscopic data alone[10,11]. Accordingly, an X-ray structure determination has been done, showing the compound to be 4,4´methylenebis(1,3,5-trimethyl-4-imidazoline-2-one) (X) in the solid state[12].

(X)



Crystal Data.

$C_{13}H_{20}N_4O_2$ (X) is monoclinic, space-group $P2_1$ (C_2^2, No. 4). a = 11.659(6), b = 10.844(47), c = 11.574(6) Å, β = 108.332(6)°, U = 1389.1 Å^3, Mr = 264.33, Dc = 1.264 g.cm^{-3}. for Z = 4. F(000) = 568.00. Mo-K_α radiation used with λ = 0.71069 Å. μ(Mo-K_α) = 0.53 cm^{-1}. The asymmetric unit consists of two molecules and all atoms occupy two-fold general positions in the unit cell.

Data Collection and Structure Solution.

The intensities of 2939 unique reflections in the quadrant $\pm$h, +k, +l, with $2^\circ \leqslant \Theta \leqslant 30^\circ$, k = 0 - 14 ($0.000^\circ \leqslant \mu \leqslant 27.308^\circ$), were measured, using a Stoe STADI-2 diffractometer with graphite monochromatised Mo-K_α radiation and a scan speed of $0.0167^\circ s^{-1}$. in ω and a scan width of 1.2° in ω.

Backgrounds were measured at each end of the scan range for a time equal to (scan time/2), and standard reflections which were measured after every fifty reflections showed only small random deviations about their means. Lorentz and polarisation corrections were applied, but no corrections for absorption were made to the data.

Systematic absences were: in h01, $l \neq 2n$; in 0k0, $k \neq 2n$, which initially suggested space-group $P2_1/c$ (C_{2h}^5, No. 14) but the space-group was found to be $P2_1$ (C_2^2, No. 4) during refinement of the structure.

The structure was solved by direct methods in $P2_1/c$ using SHELX 76[3]. The best E-map provided positions for all the non-hydrogen atoms in one molecule, showing a structure consisting of two five-membered rings, each with four substituents and joined by a single bridging atom. Isotropic refinement with carbon scattering factors assigned to each atom, enabled assignment of atoms as carbon, nitrogen or oxygen on the basis of inter-atomic distances and isotropic thermal parameters.

However, subsequent attempts at structure refinement revealed a large surplus electron density residing in one of the five-membered rings and a second molecule almost in the symmetry related position. Dropping the overall symmetry from $P2_1/c$ to $P2_1$ gave much more satisfactory results and thereafter refinement of the crystal structure was continued in the latter space-group. The second molecule of the asymmetric unit is almost, but not quite, in the symmetry related position: this pseudo-symmetry thus allowed the structure to be solved in $P2_1/c$.

Four cycles of blocked full-matrix least-squares refinement with C, N, O isotropic gave an R of 0.2131 and an Rg of 0.2288. Introduction of anisotropic thermal parameters reduced R to 0.1290 and Rg to 0.1428 for 2014 unique reflections having $F_o \geq 6\sigma'(F_o)$: in addition two very intense reflections (2,1,-1) and (0,2,0) with high D/σ' values and which were apparently affected by extinction were omitted from the data at this stage. Introduction of an $\omega^{-1} = \sigma'^2(F)$ weighting scheme caused R and Rg to increase but had no appreciable effect on the geometry of the molecule. Hence, unit weights were employed.

Introducing hydrogen atoms in calculated positions reduced R and Rg which converged to their final values of 0.1110 and 0.1164. At each stage of refinement, the decrease in Rg was significant at the 99.5 % level[7]. The change in Rg was not significant when the alternative enantiomorph was refined, because the structure is close to being centrosymmetric.

In the final cycles of refinement, 345 parameters were varied comprising 114 positional parameters, 228 anisotropic thermal parameters, two common isotropic thermal parameters (one for all CH_3- hydrogen atoms and one for the $-CH_2-$ hydrogen atoms), and one overall scale factor. Complex neutral-atom scattering factors were employed for all atoms[4,5,6].

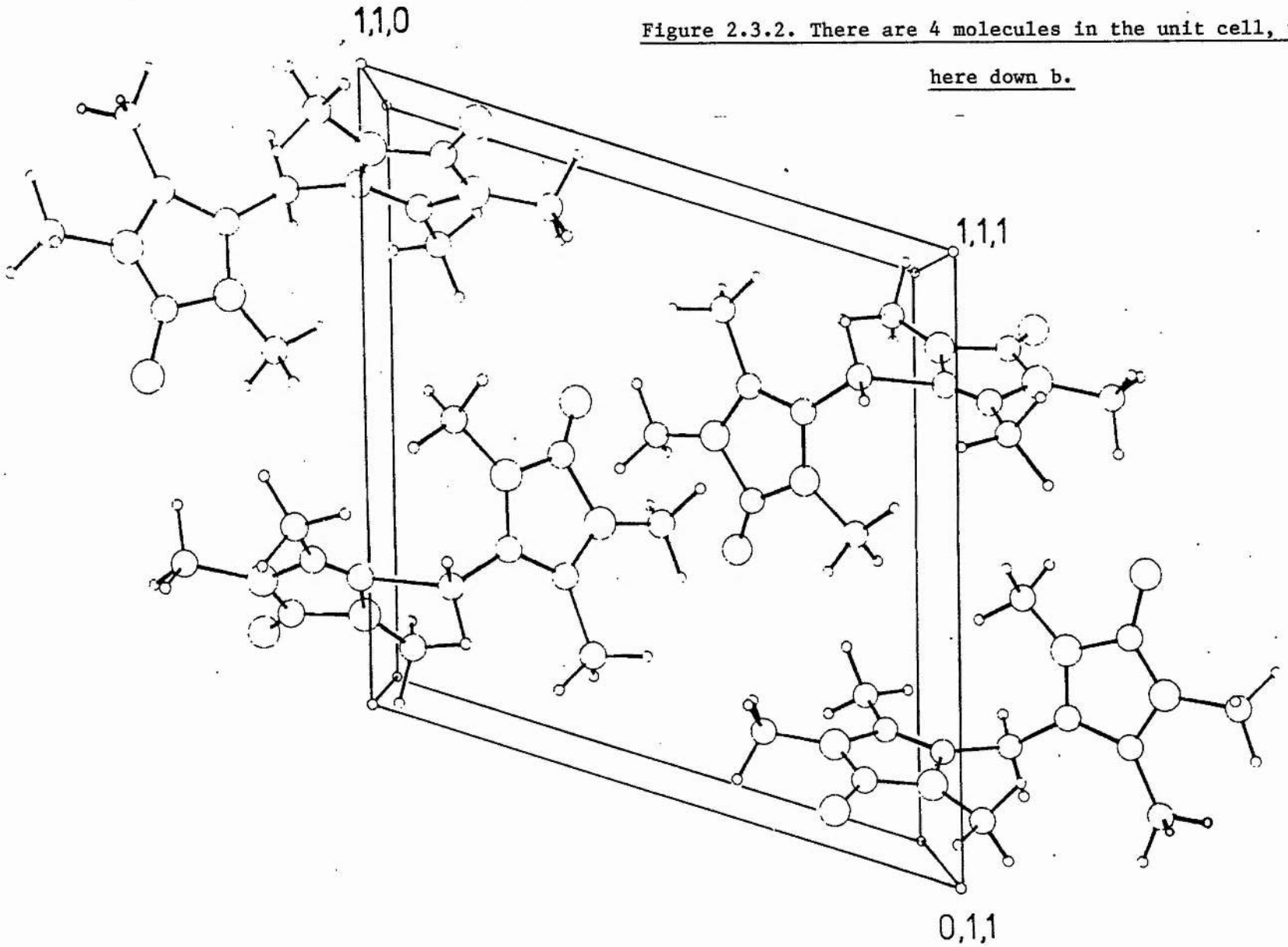


Figure 2.3.2. There are 4 molecules in the unit cell, viewed here down b.

Figure 2.3.1. The 2 molecules of the asymmetric unit.

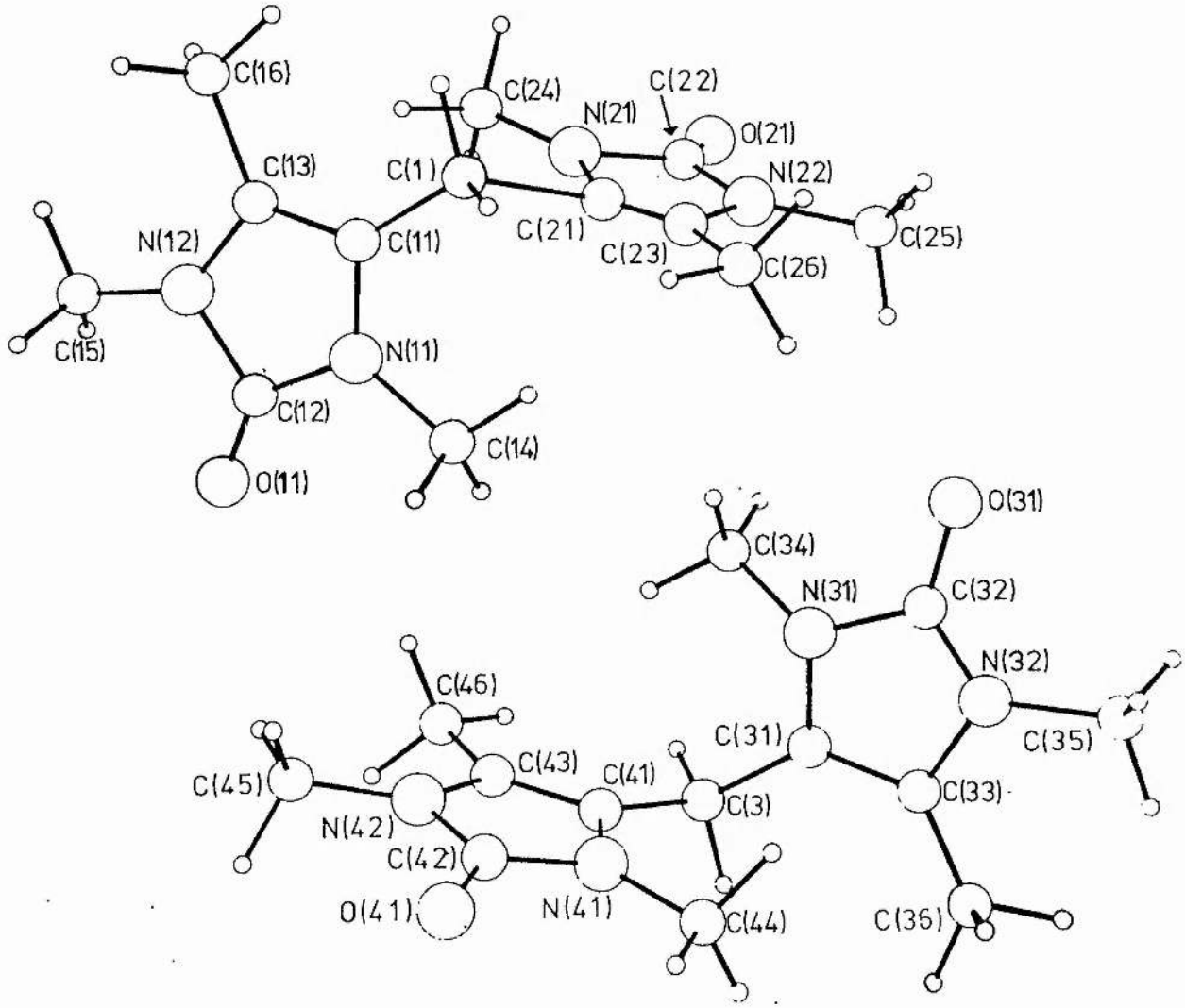


Table 2.3.1. Fractional Atomic Coordinates(x 10^4).

Atom	x	y	z
C(1)	7710(10)	5815(12)	8601(9)
O(11)	4194(9)	3414(11)	6457(9)
N(11)	5714(9)	4702811)	7653(8)
N(12)	6008(9)	3796(13)	6062(10)
C(11)	6856(10)	5103(13)	7625(9)
C(12)	5110(13)	3973(14)	6716(11)
C(13)	7016(10)	4508(13)	6677(10)
C(14)	5093(14)	5241(18)	8519(12)
C(15)	5717(13)	3099(15)	5000(11)
C(16)	8142(14)	4598(19)	6221(11)
O(21)	9508(8)	3027(10)	11975(8)
N(21)	8666(9)	4130(11)	10166(8)
N(22)	8579(8)	4981(10)	11848(7)
C(21)	8039(11)	5296(12)	9834(9)
C(22)	9027(11)	3885(17)	11438(12)
C(23)	7972(10)	5740(11)	10934(9)
C(24)	8988(12)	3238(14)	9364(11)
C(25)	8690(13)	5251(17)	13151(10)
C(26)	7510(15)	6983(16)	11153(14)
C(3)	2316(10)	4036(13)	11382(9)
O(31)	5934(8)	6243(13)	13601(9)
N(31)	4267(9)	5188(12)	12293(9)
N(32)	4043(9)	5961(11)	13926(8)
C(31)	3137(9)	4859(12)	12343(9)
C(32)	4812(11)	5885(15)	13339(11)

C(33)	2979(11)	5326(13)	13378(10)
C(34)	4911(12)	4699(18)	11574(12)
C(35)	4207(15)	6689(22)	15099(14)
C(36)	1993(10)	5314(14)	13888(11)
O(41)	436(10)	6802(12)	8025(9)
N(41)	1311(9)	5683(11)	9843(8)
N(42)	1457(10)	4978(13)	8135(8)
C(41)	1879(10)	4645(13)	10107(9)
C(42)	1050(10)	5893(12)	8605(9)
C(43)	1941(10)	4039(15)	9107(10)
C(44)	1026(12)	6523(16)	10668(12)
C(45)	1297(12)	4877(17)	6850(9)
C(46)	2381(11)	2882(12)	8813(10)

Table 2.3.2. Average Geometry of the Four Rings.

(a) Distances(Å).

C(1) - C(11)	1.499(15)	C(12) - N(12)	1.375(21)
C(11) - N(11)	1.384(17)	N(12) - C(15)	1.466(21)
N(11) - C(14)	1.454(21)	N(12) - C(13)	1.406(17)
N(11) - C(12)	1.389(17)	C(13) - C(16)	1.488(22)
C(12) - O(11)	1.231(19)	C(13) - C(11)	1.360(18)

(b) Angles(°).

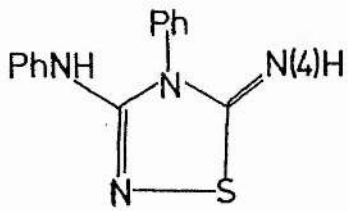
C(1) - C(11) - N(11)	123.5(11)	N(11) - C(12) - N(12)	104.3(11)
C(1) - C(11) - C(13)	128.6(12)	O(11) - C(12) - N(12)	127.8(13)
C(11) - C(1) - C(12)	115.0(11)	C(12) - N(12) - C(15)	123.5(12)
C(11) - N(11) - C(14)	126.7(12)	C(12) - N(12) - C(13)	110.4(11)

C(11) - N(11) - C(12)	110.8(11)	C(15) - N(12) - C(13)	125.9(13)
N(11) - C(11) - C(13)	107.3(10)	N(12) - C(13) - C(16)	121.5(12)
C(14) - N(11) - C(12)	121.7(11)	N(12) - C(13) - C(11)	106.8(12)
N(11) - C(12) - O(11)	127.6(15)	C(16) - C(13) - C(11)	131.3(12)

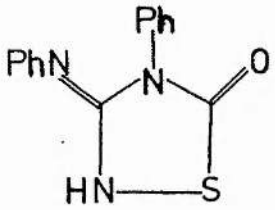
IV. 4-Phenyl-3-phenylamino-1,2,4-thiadiazoline-5-one.

Preparation.

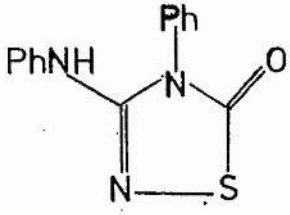
This compound is generally known as Dost's Keto Compound, and has the stoichiometry $C_{14}H_{11}N_3OS$. Oxidation of 1-phenylthiourea yields a basic compound $C_{14}H_{12}N_4S$ which is called Hector's Base[13] and whose constitution in the solid state[14] and in solution[15] is (XI). By treating Hector's Base with concentrated hydrochloric acid, a non-basic keto-compound, $C_{14}H_{11}N_3OS$, Dost's Keto Compound is obtained[16]. Although the structure (XII) has been suggested[17], our X-ray analysis shows Dost's Keto Compound to be (XIII), 4-phenyl-3-phenylamino-1,2,4-thiadiazolin-5-one. The compound was thus prepared from Hector's Base, repeatedly recrystallised from ethanol to give massive needles about 15 mm. in length. A fragment from such a crystal was used for data collection.



(XI)



(XII)



(XIII)

Crystal Data.

$C_{14}H_{11}N_3OS$ (XIII) is orthorhombic, space-group $P2_12_12_1$ (D_2^4, No. 19). $a = 12.252(9)$, $b = 11.283(8)$, $c = 9.297(10)$ Å, $U = 1285.2$ Å^3, Mr = 269.32, $Dc = 1.392$ g.cm^{-3}. for $Z = 8$. $F(000) = 560.00$. Mo-K_α radiation used with $\lambda = 0.71069$ Å, μ(Mo-K_α) = 2.01 cm^{-1}.

Data Collection and Structure Solution.

The intensities of 1231 unique reflections with $0^{\circ} \leqslant \theta \leqslant 30^{\circ}$ in the octant +h, +k, +l (layers h01 to h141, $0.000^{\circ} \leqslant \mu \leqslant 26.162^{\circ}$) were measured in the ω-2θ scan mode using a Stoe STADI-2 two-circle diffractometer with graphite monochromatised Mo-K_{α} radiation, with a scan speed of $0.0167^{\circ}s^{-1}$. in ω and a scan width of 1.2° in ω. Backgrounds were measured at each end of the scan range for a time equal to (scan time/2). After every fifty reflections standard reflections were measured and they showed only small random deviations about their means. Lorentz and polarisation corrections were applied, but no correction for absorption made.

Systematic absences were: in h00, $h \neq 2n$; in 0k0, $k \neq 2n$; in 001, $1 \neq 2n$. Hence the space-group could be determined uniquely as $P2_12_12_1$. All atoms occupy four-fold general positions and the asymmetric unit consists of only one molecule.

Dost's Keto Compound crystallises in the same space-group as Hector's Base, $P2_12_12_1$, and the unit cell dimensions are very similar. Therefore it was thought reasonable to suppose that the two compounds are isomorphous and initial coordinates were taken from those of Hector's Base with N(4) replaced by O(1).

Comparison of the Unit Cell Dimensions.

	Hector's Base[14].	Dost's Keto Compound.
Stoichiometry	$C_{14}H_{12}N_4S$	$C_{14}H_{11}N_3OS$
Space-group	$P2_12_12_1$	$P2_12_12_1$
a =	12.196(2)	12.252(9)
b =	11.027(2)	11.283(8)
c =	9.519(2)	9.297(10)
Z	4	4

Using SHELX 76[3] six cycles of refinement with individual isotropic thermal parameters and 1231 data gave R = 0.166 and Rg = 0.179 and thereafter introducing anisotropic thermal parameters for all non-hydrogen atoms and only using 1076 data having $F_o \geq 6\sigma(F_o)$ reduced Rg to 0.139.

At this stage, the difference map revealed both the position of the unique hydrogen, H(3), bonded to the exocyclic amino nitrogen N(3), and the positions of the majority of the phenyl hydrogens. Hydrogen atom coordinates were refined, starting with those from the difference map for H(3), and from calculated positions for the phenyl hydrogens, constraining the carbon - hydrogen distances to be in the range 0.96 $\pm$0.05 Å. After a further six cycles of full-matrix least-squares refinement with all hydrogens given a common isotropic thermal parameter, R converged at 0.0964 and Rg at 0.1001. At each stage of refinement the reduction in Rg was significant at the 99.5 % level[7]. The alternative enantiomorph, in an identical refinement

gave Rg = 0.1006, significantly worse at the 95 % level[7], so that this enantiomorph can be rejected.

Introduction of non-unit weights, either $\omega^{-1} = [\sigma^2(F) + K|F|^2]$ or $\omega^{-1} = \sigma^2(F)$ increased the R indices substantially: although the geometry of the molecule remained unchanged the esd's were generally increased two- or three-fold, so unit weights were used.

Complex neutral-atom scattering factors[4,5,6], were employed for all atoms, and in the final cycles of refinement 206 parameters were varied, comprising 90 positional parameters, 114 anisotropic thermal parameters, one isotropic thermal parameter and one overall scale factor. In the final cycle, all parameter shifts were less than $0.28\,\sigma$.

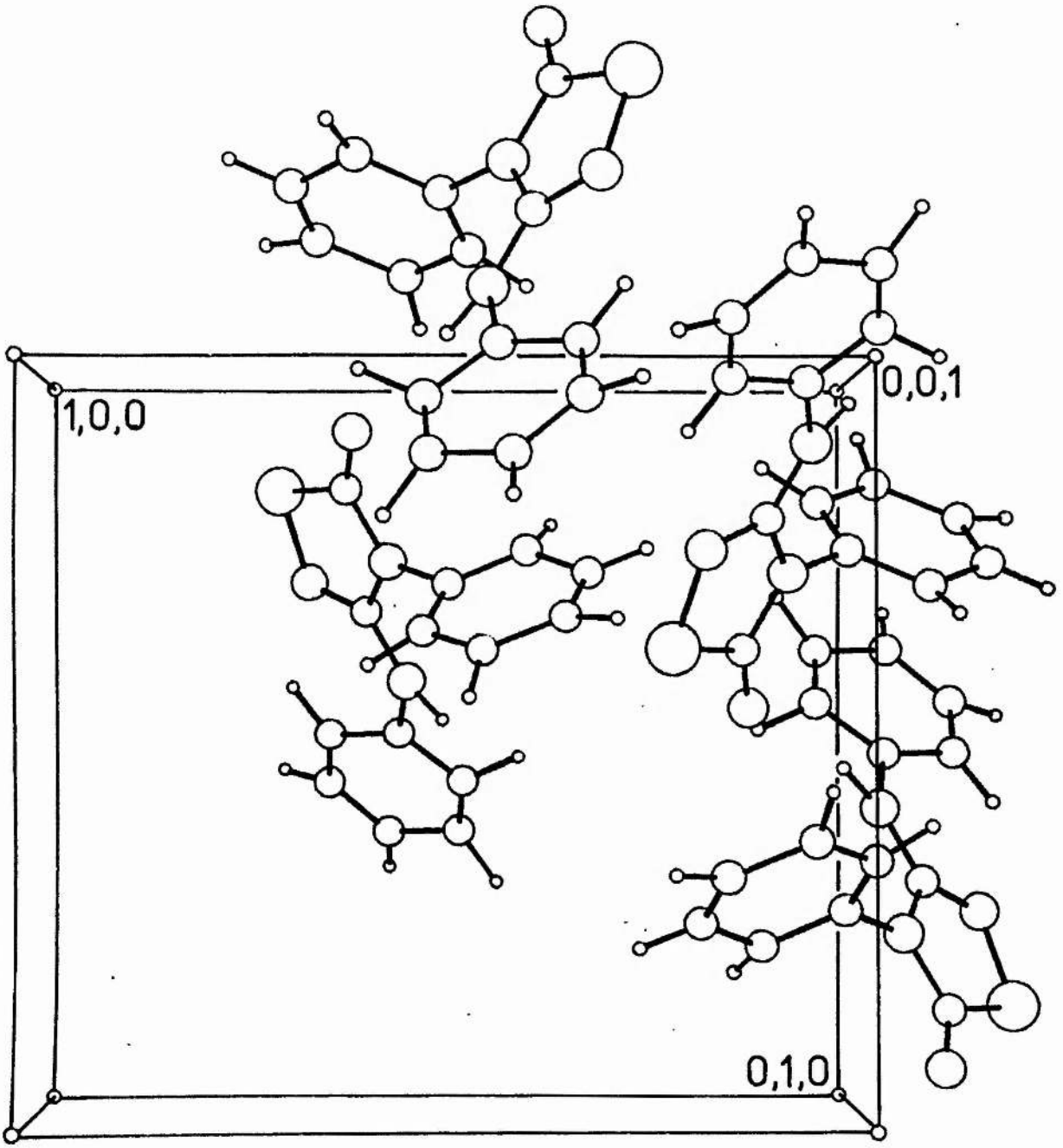


Figure 2.4.2. The contents of the unit cell, viewed down c.

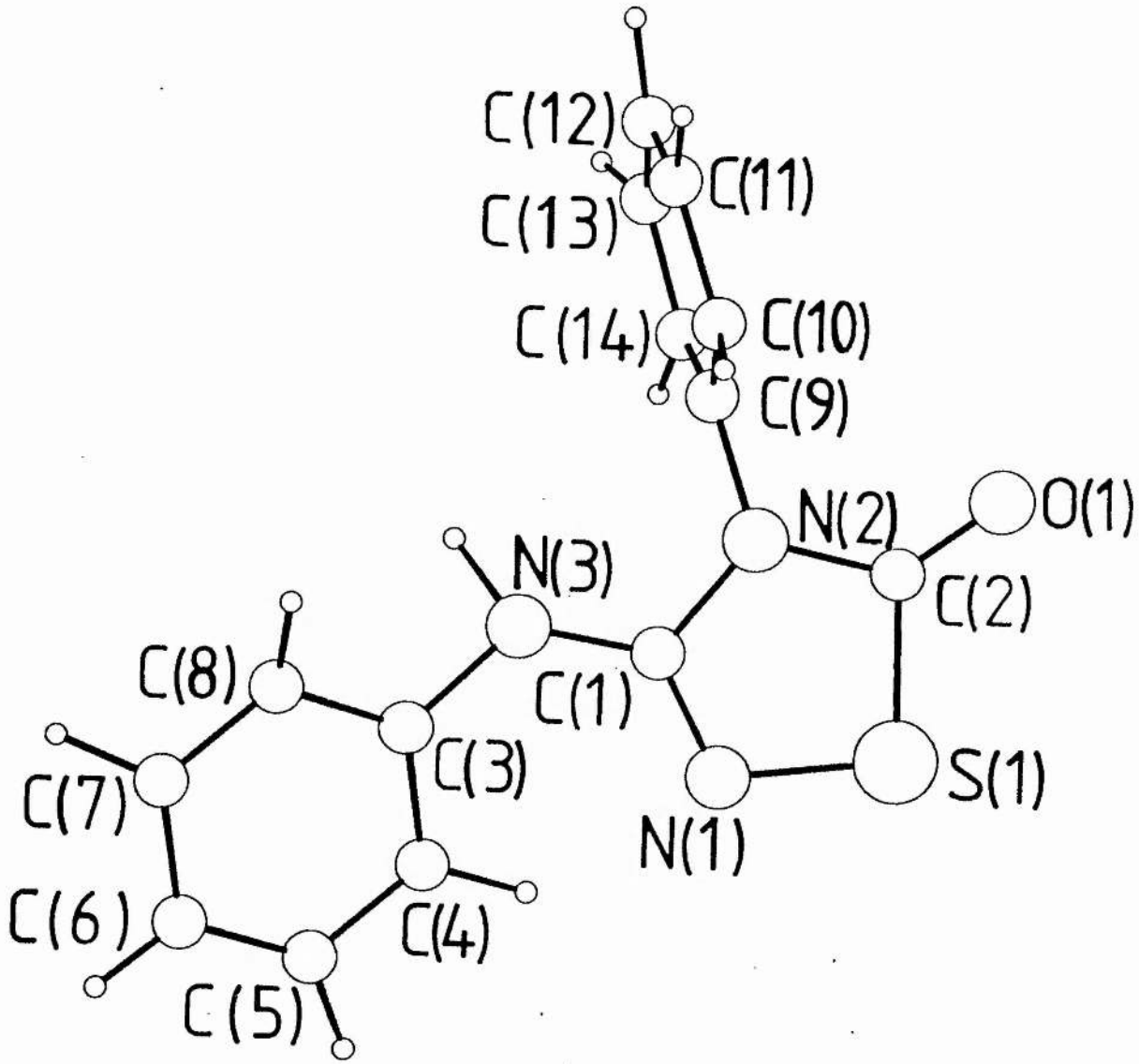


Figure 2.4.1. Dost´s Keto Compound. The molecule, showing the numbering of the atoms.

Table 2.4.1. Fractional Atomic Coordinates(x 10^4; for H x 10^3).

Atom	x	y	z
S(1)	2205(3)	3712(3)	4571(4)
N(1)	1822(8)	2382(9)	5188(4)
N(2)	753(8)	2653(8)	3181(9)
N(3)	521(8)	910(8)	4577(11)
O(1)	1024(8)	4501(8)	2187(10)
C(1)	1047(9)	1938(9)	4368(12)
C(2)	1304(10)	3707(14)	3042(14)
C(3)	627(10)	143(10)	5794(11)
C(4)	1568(11)	121(12)	6643(15)
C(5)	1596(12)	-623(15)	7801(16)
C(6)	778(14)	-1362(14)	8155(14)
C(7)	-184(13)	-1339(13)	7286(15)
C(8)	-237(12)	-579(11)	6127(14)
C(9)	-40(9)	2302(9)	2101(12)
C(10)	294(11)	1604(12)	957(14)
C(11)	-472(15)	1320(13)	-73(14)
C(12)	-1550(13)	1800(15)	50(17)
C(13)	-1847(12)	2450(16)	1178(21)
C(14)	-1067(11)	2786(14)	2215(16)
H(3)	2(11)	37(11)	394(14)
H(4)	208(8)	80(7)	650(14)
H(5)	210(9)	-27(11)	844(11)
H(6)	82(11)	-194(9)	894(9)
H(7)	-63(9)	-205(7)	737(15)
H(8)	-91(6)	-19(11)	575(14)

H(10)	102(7)	120(10)	95(14)
H(11)	-33(11)	59(7)	-63(12)
H(12)	-215(8)	190(10)	-66(11)
H(13)	-251(7)	294(10)	113(15)
H(14)	-128(10)	334(10)	303(9)

Table 2.4.2. Geometry of the Molecule.

(a) Distances(Å).

S(1) - N(1)	1.673(10)	C(9) - C(14)	1.375(16)
S(1) - C(2)	1.800(13)	C(10) - C(11)	1.378(18)
C(1) - N(1)	1.316(14)	C(11) - C(12)	1.432(21)
C(1) - N(2)	1.414(13)	C(12) - C(13)	1.331(22)
C(1) - N(3)	1.341(13)	C(13) - C(14)	1.409(20)
C(2) - O(1)	1.204(17)	N(3) - H(3)	1.05(13)
C(2) - N(2)	1.374(17)	C(4) - H(4)	1.00(5)
C(3) - N(3)	1.431(14)	C(5) - H(5)	0.94(5)
C(3) - C(4)	1.398(16)	C(6) - H(6)	0.98(5)
C(3) - C(8)	1.371(17)	C(7) - H(7)	0.97(5)
C(4) - C(5)	1.366(18)	C(8) - H(8)	1.00(5)
C(5) - C(6)	1.344(21)	C(10) - H(10)	1.00(5)
C(6) - C(7)	1.429(19)	C(11) - H(11)	0.98(5)
C(7) - C(8)	1.379(18)	C(12) - H(12)	1.00(5)
C(9) - N(2)	1.452(13)	C(13) - H(13)	0.98(5)
C(9) - C(10)	1.385(16)	C(14) - H(14)	1.02(5)

(b) Angles(°).

C(1) - N(1) - S(1)	110.2(8)	N(3) - C(3) - C(8)	117.9(11)

C(1)	- N(2)	- C(2)	116.3(10)	C(4)	- C(3)	- C(8)	119.9(11)
C(2)	- S(1)	- N(1)	95.5(6)	C(3)	- C(4)	- C(5)	118.5(13)
N(1)	- C(1)	- N(2)	114.7(10)	C(4)	- C(5)	- C(6)	123.7(14)
N(2)	- C(2)	- S(1)	103.3(10)	C(5)	- C(6)	- C(7)	117.7(12)
O(1)	- C(2)	- S(1)	125.6(11)	C(6)	- C(7)	- C(8)	119.5(13)
O(1)	- C(2)	- N(2)	131.1(12)	C(7)	- C(8)	- C(3)	120.6(13)
C(1)	- N(2)	- C(9)	123.7(9)	C(9)	- C(10)	- C(11)	117.7(13)
C(2)	- N(2)	- C(9)	120.0(10)	C(10)	- C(11)	- C(12)	119.0(13)
N(1)	- C(1)	- N(3)	126.3(11)	C(11)	- C(12)	- C(13)	121.5(14)
N(2)	- C(1)	- N(3)	119.0(10)	C(12)	- C(13)	- C(14)	120.1(15)
N(2)	- C(9)	- C(10)	119.3(10)	C(13)	- C(14)	- C(9)	117.5(14)
N(2)	- C(9)	- C(14)	116.8(11)	C(14)	- C(9)	- C(10)	123.7(12)
C(1)	- N(3)	- C(3)	126.5(10)	C(1)	- N(3)	- H(3)	135(7)
N(3)	- C(3)	- C(4)	122.2(11)	C(3)	- N(3)	- H(3)	98(7)

Chapter Two.

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Chapter Three.

The Four Heterocyclic molecules and related compounds.

I. Trithiapentalenes and linear multisulphur systems.

The structure of 6a-trithiapentalene, which is almost planar with C_{2v} symmetry is shown below, along with many substituted derivatives which have been investigated crystallographically. If one calculates the overall distance for the S-S-S skeleton, it is remarkably constant, being approximately 4.65 to 4.74 Å , with the three sulphur atoms almost linear (hence the simple summation of S - S distances).

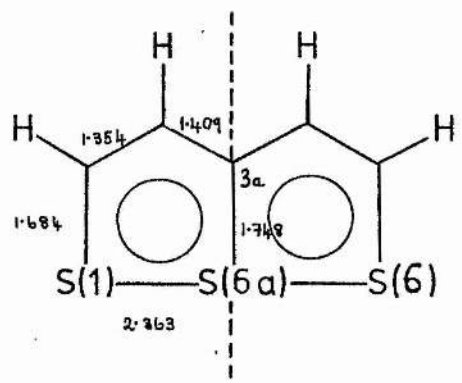
However, different substituents in various orientations profoundly affect the individual bond lengths (please see Table 3.1) whilst the overall distance, as mentioned, remains constant.

Intuitively, symmetrical derivatives might be expected to have a symmetrical S-S-S skeleton. Whilst (I) and (II) are symmetric, it is obvious that (III) and (IV) are asymmetric. In (III) the phenyl orientations are very different; however, the two phenyl groups in (IV) are roughly parallel - and yet have S - S distances so very different. (V) has very different substituents, but the S - S distances are almost the same.

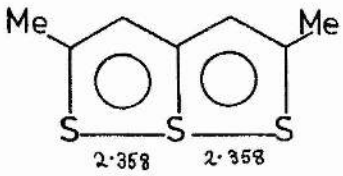
Table 3.1. S(1) - S(6a) and S(6a) - S(6) Distances (Å).

Compound	S(1) - S(6a)	S(6a) - S(6)	S...S	Ref.
I	2.363(1)	2.363(1)	4.726(1)	1
II	2.358(1)	2.358(1)	4.716(1)	2
III	2.362(3)	2.304(3)	4.666(3)	3
IV	2.232(4)	2.434(4)	4.666(4)	4
V	2.348(2)	2.350(2)	4.698(2)	5
VI	2.431(2)	2.308(2)	4.739(2)	6
VII	2.481(2)	2.242(2)	4.723(2)	7
VIII	2.255(1)	2.398(1)	4.653(1)	8
IX	2.351(1)	2.351(1)	non linear	9
X	2.329(1)	2.288(1)	4.617	10

Figure 3.1. 6a-Trithiapentalenes.



(I)



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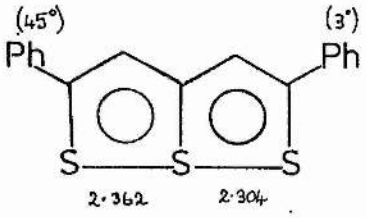

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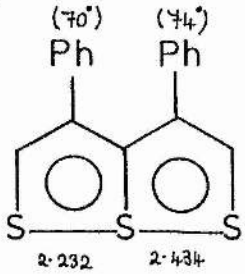

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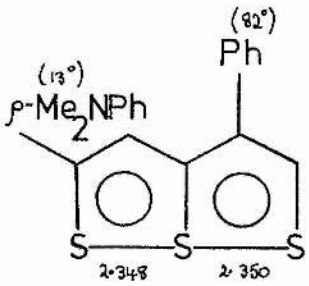

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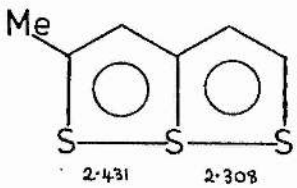
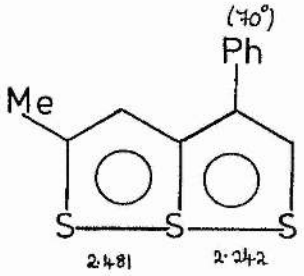


(VI)



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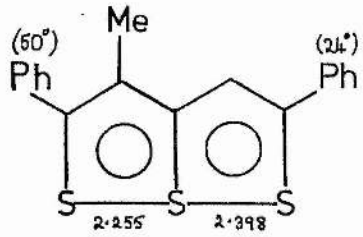
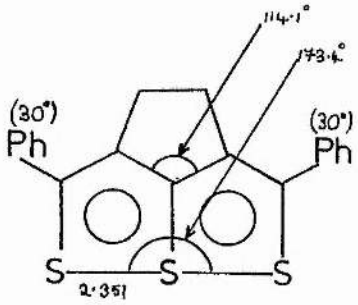


(VIII)



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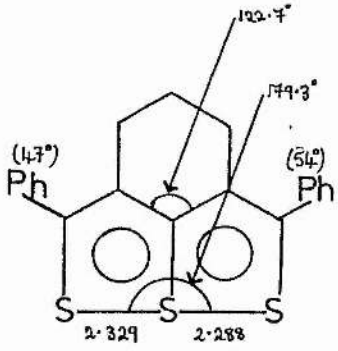
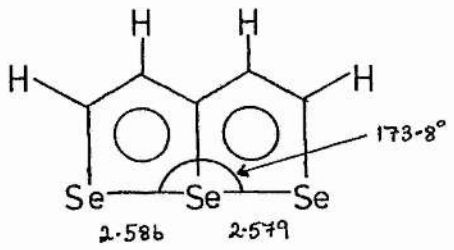


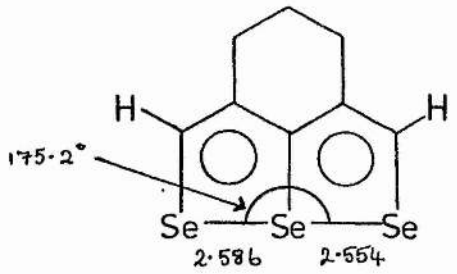
(X)

CNDO calculations[11], carried out by Hørdvik et al., for simple 6a-trithiapentalenes in which the overall S(1)...S(6) distance was kept constant, but the position of S(6a) was varied about the central point, gave a potential energy curve with a broad, flat minimum for the parent compound, (I). The width of this broad, flat potential energy curve was ca. 0.3 Å: thus S(6a) may move 0.15 Å to either side of the central point and the potential energy of the molecule changes only negligibly. This may show that only small energy changes are involved for different S - S bond lengths - and that different electron donating or withdrawing ability of the substituents will change the position of S(6a). Asymmetric Me derivatives are calculated to have asymmetric S-S-S with Me in the 2-position lengthening the S(1) - S(6a) distance and in the 3-position shortening it. Structures (VI), (VII) and (VIII) agree with this although the effect of the various orientations of the phenyl groups is difficult to assess. A suggestion that not only do the substituents affect S(6a)'s position intramolecularly, but also that intermolecular forces in the crystal are important, may be made, since (IV) is so very asymmetric whilst the phenyl groups' orientations are alike.

Compounds (IX) and (X) are of interest because a 3,4-di or trimethylene bridge either shortens the S-S-S distance to ca. 4.61 Å as in (X), or the strain deforms the S-S-S skeleton from approximate linearity, (IX). A similar shortening effect caused by a trimethylene bridge may be observed in the 6a-triselenapentalenes. Compare the Se - Se distances in (XI)[12] and (XII)[13].



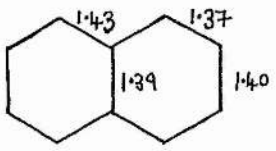
(XI)



(XII)

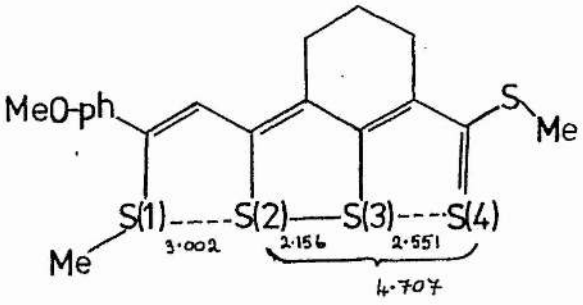
The trithiapentalenes and other Group VI analogues have been represented as a delocalised bicyclic system, although as in naphthalene[14] C - C bond lengths differ in these rings.

Bond lengths in Naphthalene

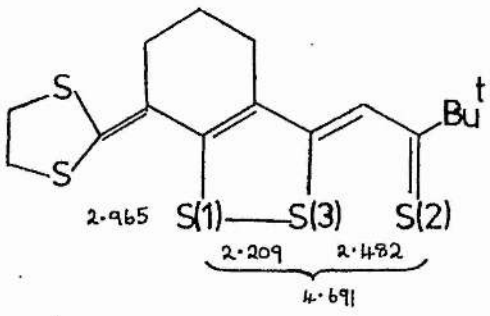


The S-S-S skeleton is regarded as a 3 centre, 4 electron bond. Other collinear multisulphur systems with approximate planarity have been investigated, and immediately pose the problem of which is the best way to represent their structure.

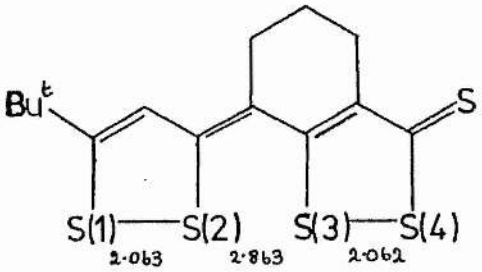
Some tetra-sulphur compounds have been represented as



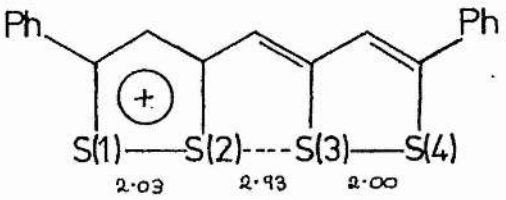
(XIII(i))[15]



(XIV(i))[15,16]



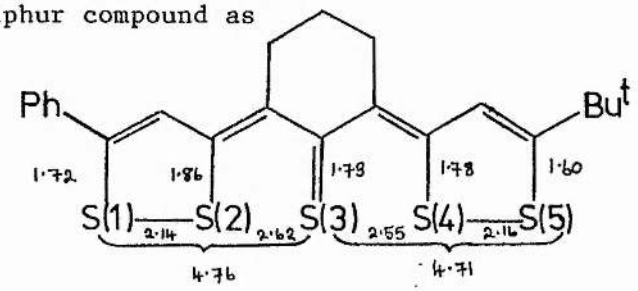
(XV)[15,16]



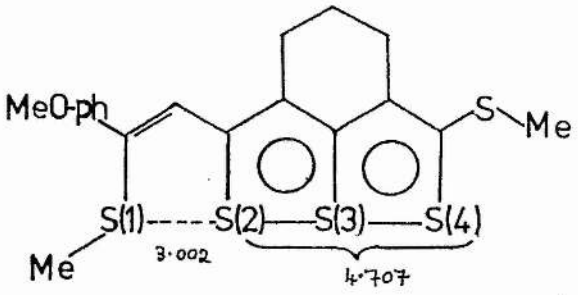
(XVI)[15,16]

and a penta-sulphur compound as

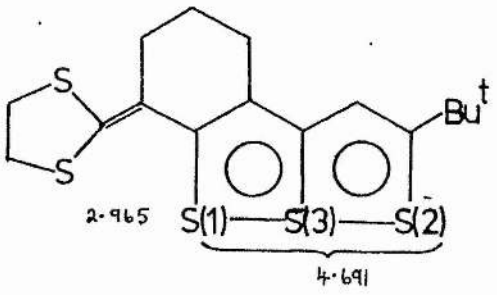
(XVII(i))[17]



In the light of the previous way of describing the S-S-S skeleton as a 3 centre, 4 electron bond, would not (XIII(i)), (XIV(i)) and (XVII(i)) be better represented as having their S(3)'s hypervalent? Thus:

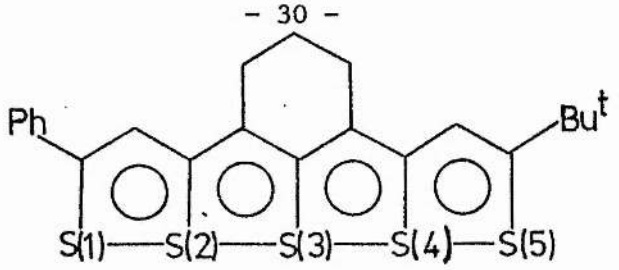


(XIII(ii))



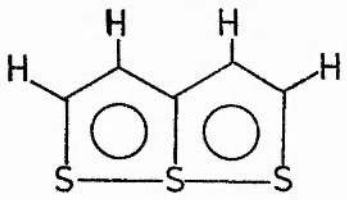
(XIV(ii))

(XVII(ii)



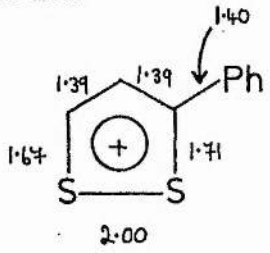
since the overall S-S-S distances in the cyclic systems sum to approximately the 4.6 - 4.7 Å which was so constant in the trithiapentalene derivatives. Representing doubly bonded S(3) in (XVII(i)) does not quite fit the data for the S - C bond lengths for S(1) and S(5), shown formally as single bonds but which are indeed shorter! This is also true of 6a-trithiapentalene (I). The S(6a) - C(3a) distance of 1.748 Å, is longer than the S - C bonding distances for S(1) and S(6), 1.684 Å. (I) has also been represented in this way:

(I)



A delocalised π-bonding pattern should therefore be envisaged in these types of compounds - the approximate planarity will also lend itself to π delocalisation. But, when is a partial bond not really a bond at all, with an internuclear distance representing only small electron interaction? Comparing the geometry of (XVI(i)) with that of the simple 3-phenyldithiolium ion[18]

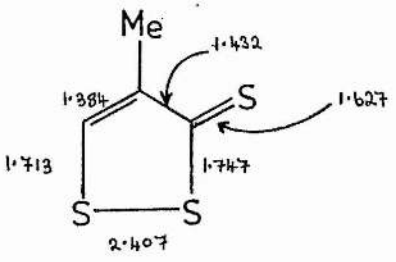
(XVIII)



would indicate that (XVI(i)) is a derivative of this cation and that

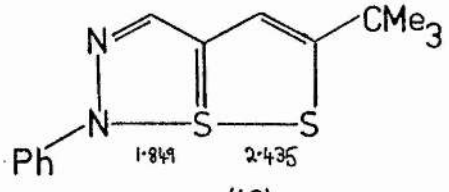
the S(2)....S(3) distance of 2.93 Å is too long for significant interaction, as is that of 2.863 Å in (XV(i)) (whose S - S distance is comparable to that of (XIX)[18]) and 3.002 Å between S(1) and S(2) in (XIII(ii)), to occur.

(XIX)

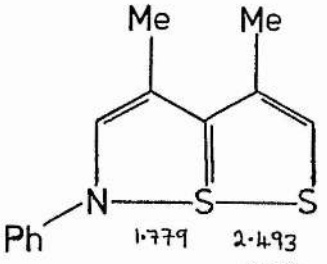


II. 5,6-Dihydro-3-thiobenzoylmethylene-3H-thiazolo[2,3-c][1,2,4] thiadiazole and related compounds.

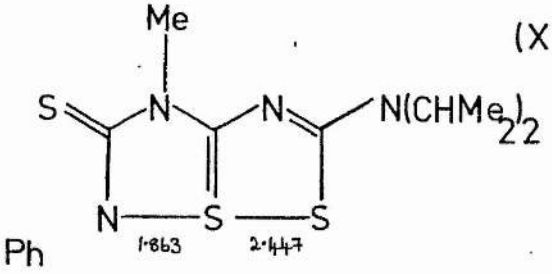
Several triheterapentalenes, analogous to the trithiapentalenes but having an N-S-S skeleton have been investigated, for example (XX) to (XXII).



(XX)[19]

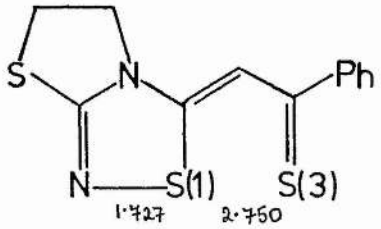


(XXI)[20]

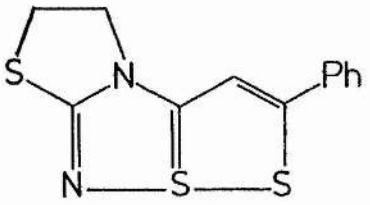


(XXII)[21]

The structure determination was carried out to find whether the title compound is best represented as (XXIII) where the S - S distance is longer and interaction less, or as (XXIV) with a hypervalent sulphur atom.



or



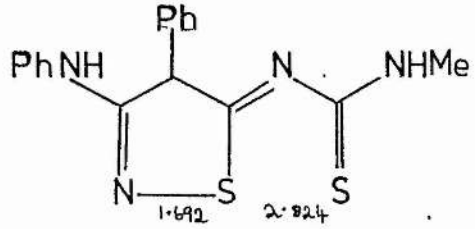
(XXIII) (XXIV)

The S - N distance 1.727(10) Å and long S - S distance 2.750(5) Å indicate the former. In structures (XX) to (XXIII) the N - S distances 1.849, 1.779, 1.863 Å are longer than the 1.727 Å we found, whilst the S - S distances of 2.435, 2.493, 2.447 Å are shorter than our 2.750 Å. On this basis, only negligible interaction between S(1) and S(3) is suggested, there being no formal bonding and hence no extensive π electron delocalisation over a bicyclic system.

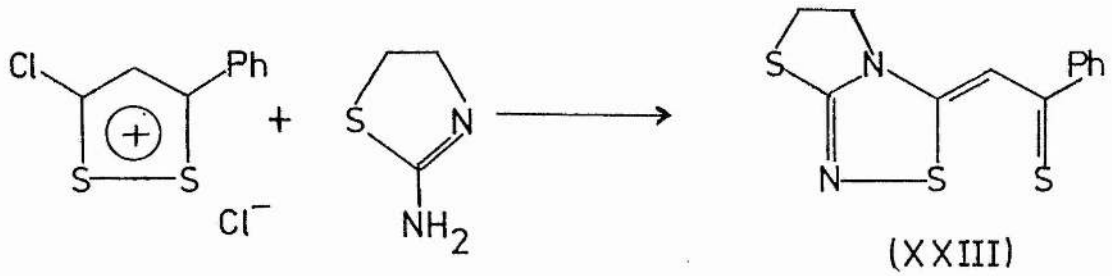
Comparing the overall N....S distances in (XX) to (XXII) shows them to be very constant (cf. the S-S-S distances previously discussed) being 4.273, 4.267 and 4.290 Å respectively[19 - 21], and shorter by ca. 0.2 Å than in the title compound, (4.471 Å),[22] and others which appear to have very similar skeletons such as (XXV). This is the methyl isothiocyanate adduct of Hector's Base[23]. In general N....S distances in skeletons of the type N-S-S seem to be approximately 4.2 Å, whilst the overall N....S distance in a skeleton

of the type N-S...S is nearer to 4.5 Å.

(XXV)



Using the criteria of interatomic distances outlined above, (XXIII) is the favoured representation of the product of the reaction of 3-chloro-5-phenyl-1,2-dithiol-1-ium chloride with 2-amino-4,5-dihydrothiazole.



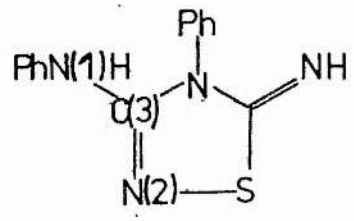
(XXIII)

Since S - S bonding occurs in the dithiolium salt, and N - S bonding distances, with long S....S distances in the product, the reaction must involve a 'bond switch' mechanism[24].

III. Analogues of Hector's Base.

Recent X-ray work by Butler, Glidewell and Liles[25] has shown the solid-state structure of Hector's Base, a heterocyclic compound originally prepared by D.S. Hector in 1889[26], to be (XXVI(i)).

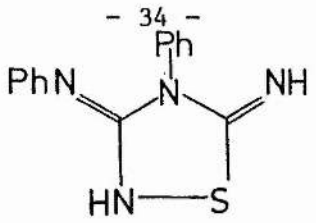
(XXVI(i))



C(3)-N(1) 1·365(4)

N(2)-C(3) 1·294(4)

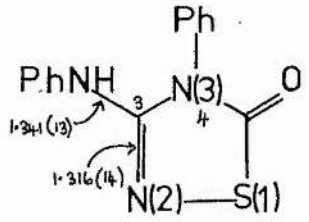
(XXVI(ii)



Until this work, a definitive structure for Hector's Base was not known, although (XVI(ii)) was a favourite contender. This representation is still favoured by Akiba in spite of the short N(2) - C(3), 1.294(4) Å, and long N(1) - C(3), 1.365(4) Å.

Treatment of Hector's Base with concentrated hydrochloric acid gives Dost's Keto Compound (XXVII) whose solid-state structure[27] was reported earlier. Again, until interatomic distances were found, it was not known whether a phenylamino or phenylimino group is present in the 3-position. The compounds are isomorphous ($P2_12_12_1$) and isostructural, the 5-imino group of Hector's Base having been converted to a carbonyl function. Dost's Keto Compound may be represented as (XXVII).

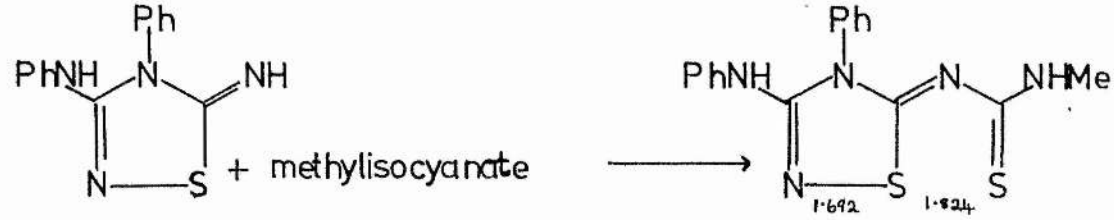
(XXVII)



The molecule is planar with a delocalised π electron sextet: N(2) and C(3) each providing one electron and S(1) and N(3), in the reduced 4-position, electron pairs.

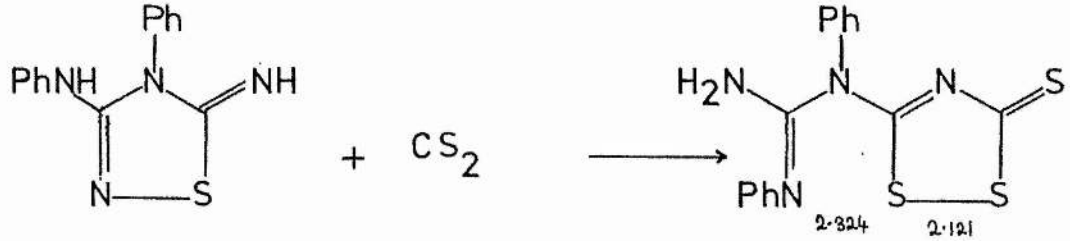
The adduct of Hector's Base with methyl isothiocyanate (XXV) has been mentioned: other 1:1 adducts have been investigated, and show that prediction of which product is obtained is difficult.

(i)



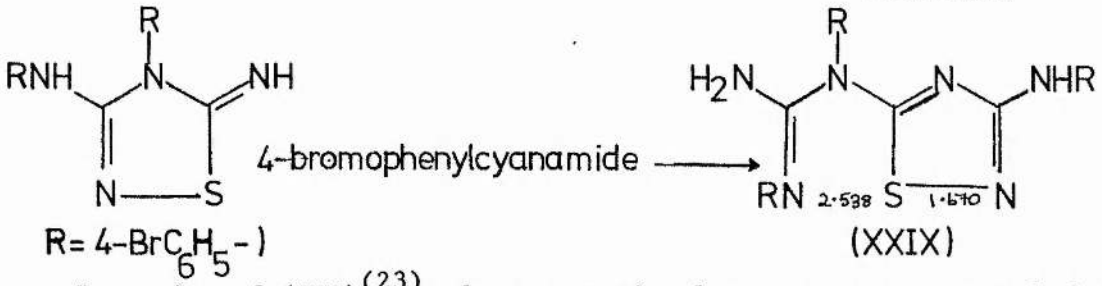
(XXV)

(ii)



(iii)

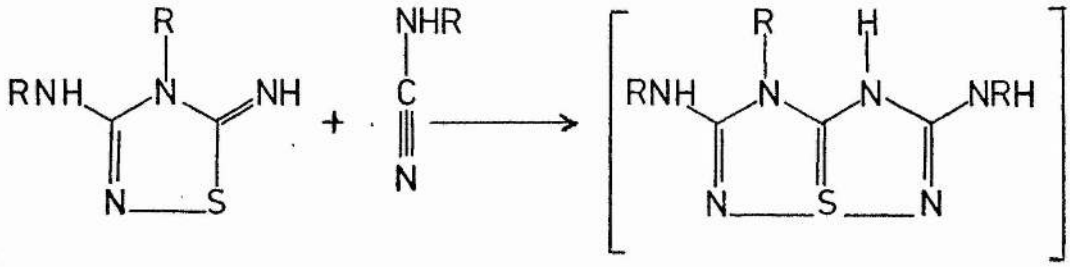
(XXVIII)

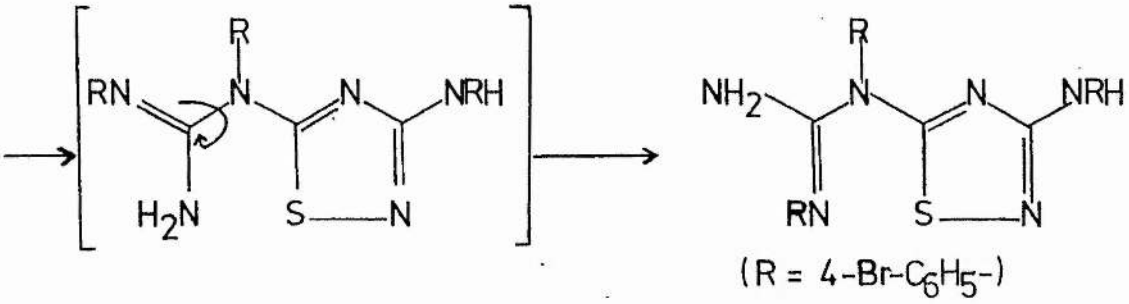


R= 4-BrC6H5-)

(XXIX)

Formation of (XXV)[23] does not involve rearrangement of the skeleton of Hector's Base, whilst (XXVIII)[28] and (XXIX)[29] clearly do, and must involve a 'bond switch', followed by prototropy and then bond rotation. A mechanism for the formation of (XXIX) and hence for this 'bond switch' type of reaction, has been suggested by Mitchell[30]:





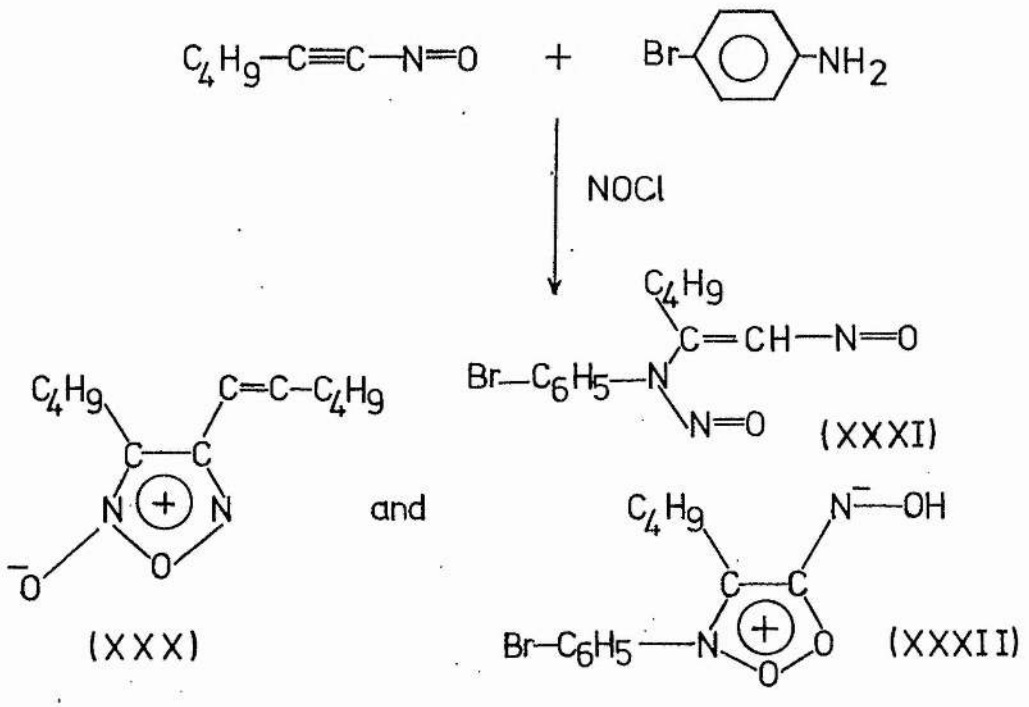
(R = 4-Br-C_6H_5-)

These adducts are found to be virtually planar and all are 10π electron systems. So is (XXIII): although the pattern of the N-S-S skeleton bonding in (XXVIII) is opposite to that of (XXIII) where the N-S...S distance was about 4.5 Å, the overall distance in (XXVIII) of about 4.44 Å is strikingly similar.

Depending on whether in a skeleton XYZ (viz. SSS, SSN, NSN, etc.) there is formally a hypervalent central atom or not, the overall X....Z distance is remarkably constant for a given set of XYZ atoms. It is atom Y which seems to move position readily: the substituents in the pseudo-pentalene skeleton affect its position, in some compounds so much that there is no formal bonding and the X....Z distance increases slightly, but noticeably, in a way not yet investigated. Possible factors are steric effects or electron withdrawing/donating ability. Dependence of position at which the substituent is found has already been mentioned for trithiapentalenes, so a similar situation may be expected for triheterapentalenes and related compounds.

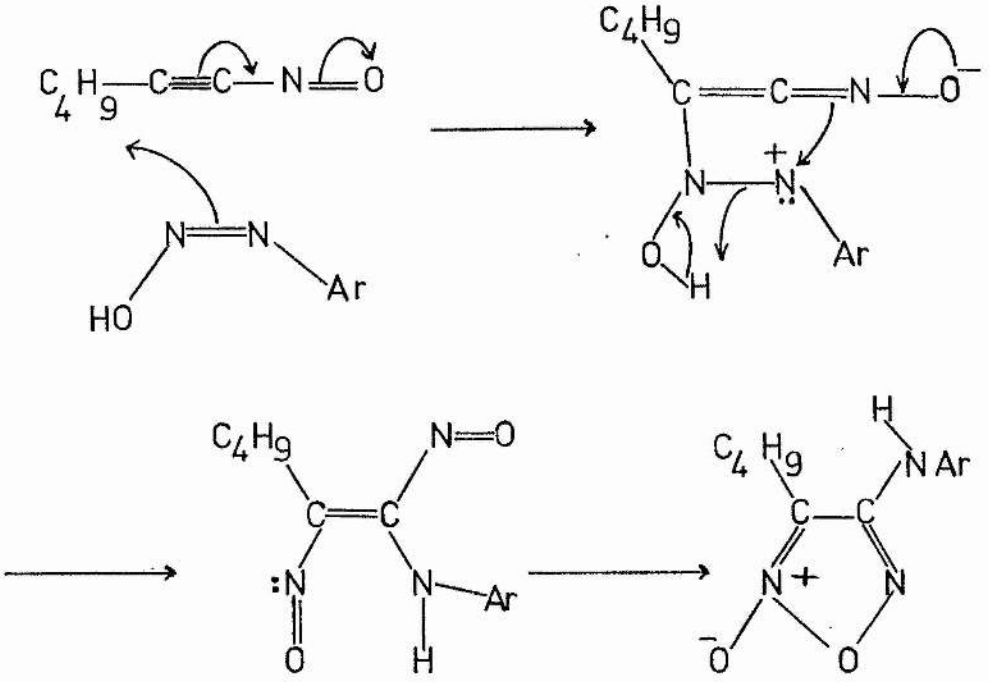
IV. A Furoxan, or a Sydnone Oxime?

By treating a solution of 1-nitroso-hex-1-yne with 4-bromoaniline and nitrosyl chloride, two compounds are formed[31]. The first is a dimer, 3-butyl-4-hex-1-ynyl-furoxan (XXX) and the second, having stoichiometry $C_{12}H_{14}BrN_3O_2$ was originally thought to be the open chain compound (XXXI). This assignment fitted well with mass spectral analysis and microanalysis, but infra-red spectra suggested either an -NH or -OH group could be present. Another possibility was that the compound was a sydnone oxime (XXXII) but this did not fit in with the fragmentation pattern in the mass spectrum, which indicated loss of two NO moieties. An X-ray structure determination[32] showed that the second compound is 4-(4-bromophenylamino)-3-butyl-furoxan, (XXXIII), which in retrospect corresponds to all spectral data. A possible reaction mechanism has been suggested by Tedder and Woodcock[31] and, knowing that the second product is indeed (XXXIII), formation of (XXXIV) by hydrogenation is very readily explained.



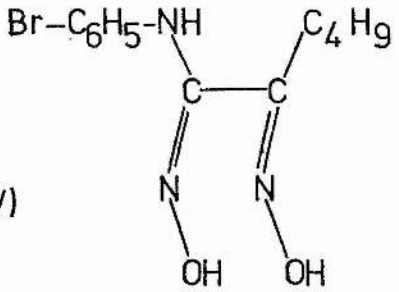
$$2\,ArNH_2 + NOCl \longrightarrow ArN_2OH + ArNH_3^+\,Cl^-$$

(Ar = 4-BrC_6H_5-)



(XXXIII)

hydrogenation

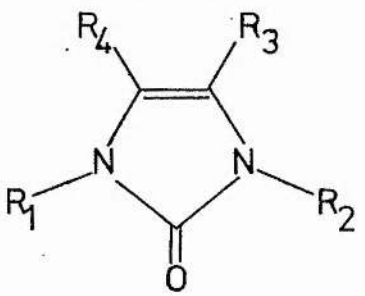


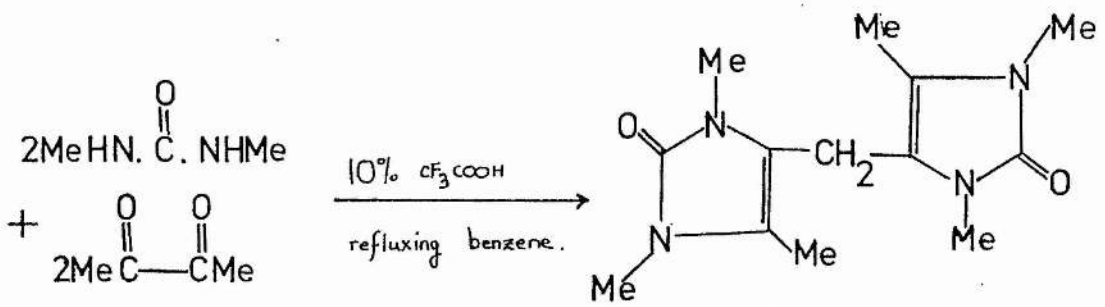
(XXXIV)

V. An unexpected reaction of 1,3-dimethylurea.

During a series of reactions of methylsubstituted ureas with various acyloins in acid medium, Butler and Hussein obtained an unexpected product when 1,3-dimethylurea reacted with butan-2,3-dione[33]. Acyloins usually react with ureas under such conditions to give 4-imidazolin-2-ones (XXXV): the unusual product, however, had stoichiometry $C_{13}H_{20}N_4O_2$ and must have been formed by reaction of two molecules of each of the reactants, loss of 4 equivalents of water and one carbon atom. Normal ^{1}H and ^{13}C n.m.r. techniques showed unexpectedly simple spectra, indicating two fold molecular symmetry, and an infra-red spectrum indicates the presence of carbonyl groups and carbon-carbon double bonds. But a definitive structure could not be assigned. Accordingly, an X-ray structure determination was carried out[34] and this compound is found to be 4,4'-methylenebis(1,3,5-trimethyl-4-imidazolin-2-one), (XXXVI).

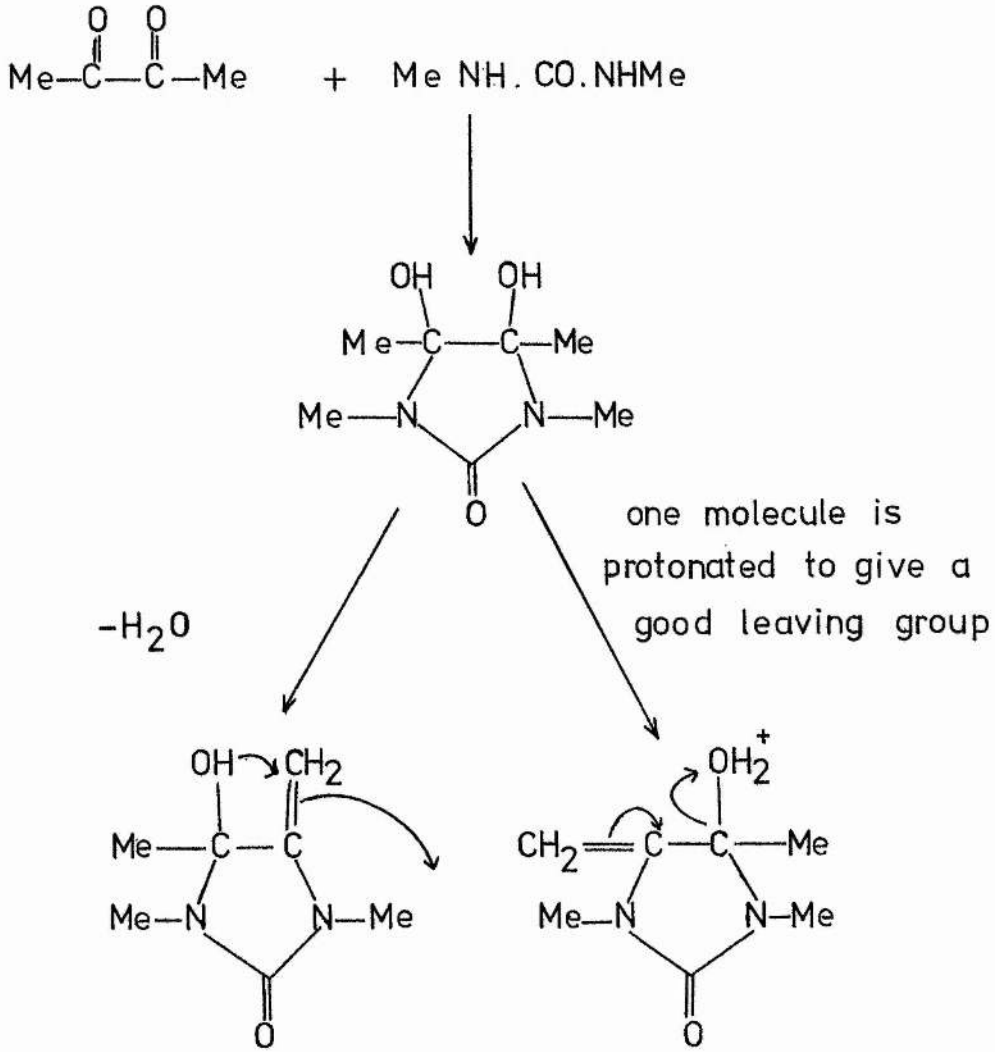
(XXXV)

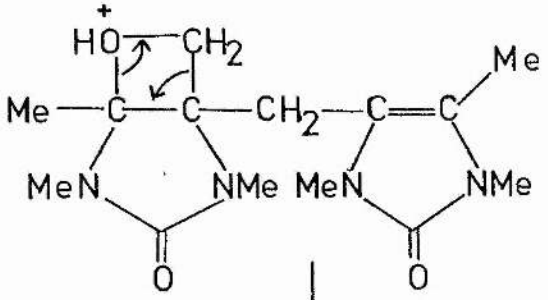


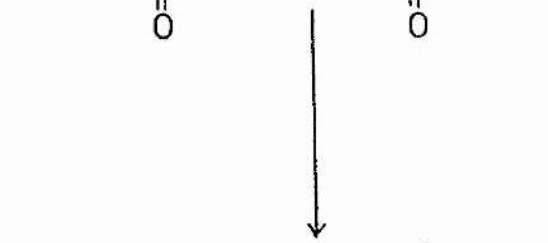


(XXXVI)

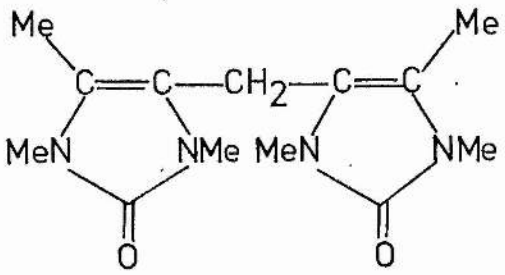
Although the molecule is not strictly symmetrical in the crystal and the asymmetric unit consists of two almost, but not absolutely, identical molecules, in solution all molecules will be equivalent and each half of a molecule will be identical thus giving rise to the extremely simple n.m.r. spectra. Following the X-ray work, a possible mechanism for this reaction was suggested by Butler and Hussein[33].







(XXXVI)



+ CH_2O + H^+

Chapter Three.

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Chapter Four

Two Compounds which contain Bis-cations: Their structural investigation by X-ray crystallography.

I. Bis(4-methylpyridine)hydrogen(I) tetraphenylborate.

Preparation.

This compound was prepared by bubbling dry hydrogen chloride through freshly distilled 4-methylpyridine to form 4-methylpyridinium chloride, a white compound which was then dried. 0.30g. of 4-methylpyridinium chloride were then dissolved in 15ml. water and a solution of 0.68g. $NaBPh_4$ in 30ml. 1:1 4-methylpyridine and acetonitrile then added; glistening needles crystallised from the solution, which were directly suitable for X-ray work[1].

Crystal Data.

$C_{36}H_{35}N_2B$ is monoclinic, space-group C2/c (C_6^{2h}, No.15). a = 16.095(14), b = 10.492(19), c = 16.763(21) Å, β = 108.90(2)°, U = 2678.2 Å^3. M_r = 506.50, D_c = 1.25 g.cm.$^{-3}$ for Z = 4. F(000) = 1080.00. Mo-K_α radiation used with λ = 0.71069 Å, μ(Mo-K_α) = 0.37cm^{-1}.

Data Collection and Structure Solution.

The intensities of 4848 reflections having $2^{\circ} \leqslant \Theta \leqslant 30^{\circ}$, in the quadrant $\pm h$, $+k$, $+l$, ($l = 0 - 11$, $0^{\circ} \leqslant \mu \leqslant 21.872^{\circ}$), were measured, using a Stoe STADI-2 diffractometer in the $\omega - 2\Theta$ scan mode with a step width of 0.01° in ω, a step time of 0.6s. and 120 steps per peak scan. At each end of the peak scan, backgrounds were measured for a time of (scan time/2). Standard reflections, which were measured every fifty reflections, showed only small random deviations about their means. No corrections for absorption were made to the data, although Lorentz and polarisation corrections were applied.

Systematic absences: hkl, $h+k \neq 2n$, h0l, $l \neq 2n$ suggested space-groups Cc or C2/c. The high E statistics indicated centrosymmetry and the following structure solution and refinement indeed confirmed C2/c. Hence the asymmetric unit consists of one half of a formula unit.

The structure was solved by direct methods in C2/c using the SHELX-76 programme system[2]. The best E-map found all non-hydrogen atoms in the asymmetric unit, to reveal the cation lying across a centre of inversion, and the anion on a twofold axis, with the boron atom occupying the special position 4e of symmetry 2 and having a site occupation factor of 0.5.

Blocked full-matrix least-squares refinement with complex neutral atom scattering factors[3,4] and C,B,N atoms anisotropic gave $R(= \Sigma\Delta/\Sigma|F_o|$ where $\Delta = |F_o - F_c|)$ = 0.1191 and $Rg[=(\Sigma\omega\Delta^2/\Sigma\omega|F_o|^2)^{1/2}]$ = 0.1189 for 1004 unique reflections having $F_o \geq 6\sigma(F_o)$.

At this stage electron density peaks were located in the difference map for the aromatic hydrogens and also for the unique hydrogen lying exactly on the centre of inversion between the two nitrogens of the bis(4-methylpyridine)hydrogen(I) cation occupying the site 4d, with fractional atomic coordinates of 1/4, 3/4, 1.

Isotropic refinement of these hydrogen atom positions, with separate thermal parameters for the unique hydrogen, the pyridine hydrogens and the phenyl hydrogens, together with the inclusion of the methyl hydrogens in calculated positions with d(C-H) = 1.08 Å and a fourth isotropic thermal parameter for these, led to a final R of 0.0785 and Rg of 0.0755. The final difference map showed no electron density > 0.2 e $Å^{-3}$.

In the final cycles of refinement, 223 parameters were varied, comprising 100 positional coordinates, 118 anisotropic thermal parameters, 4 isotropic thermal parameters and an overall scale factor.

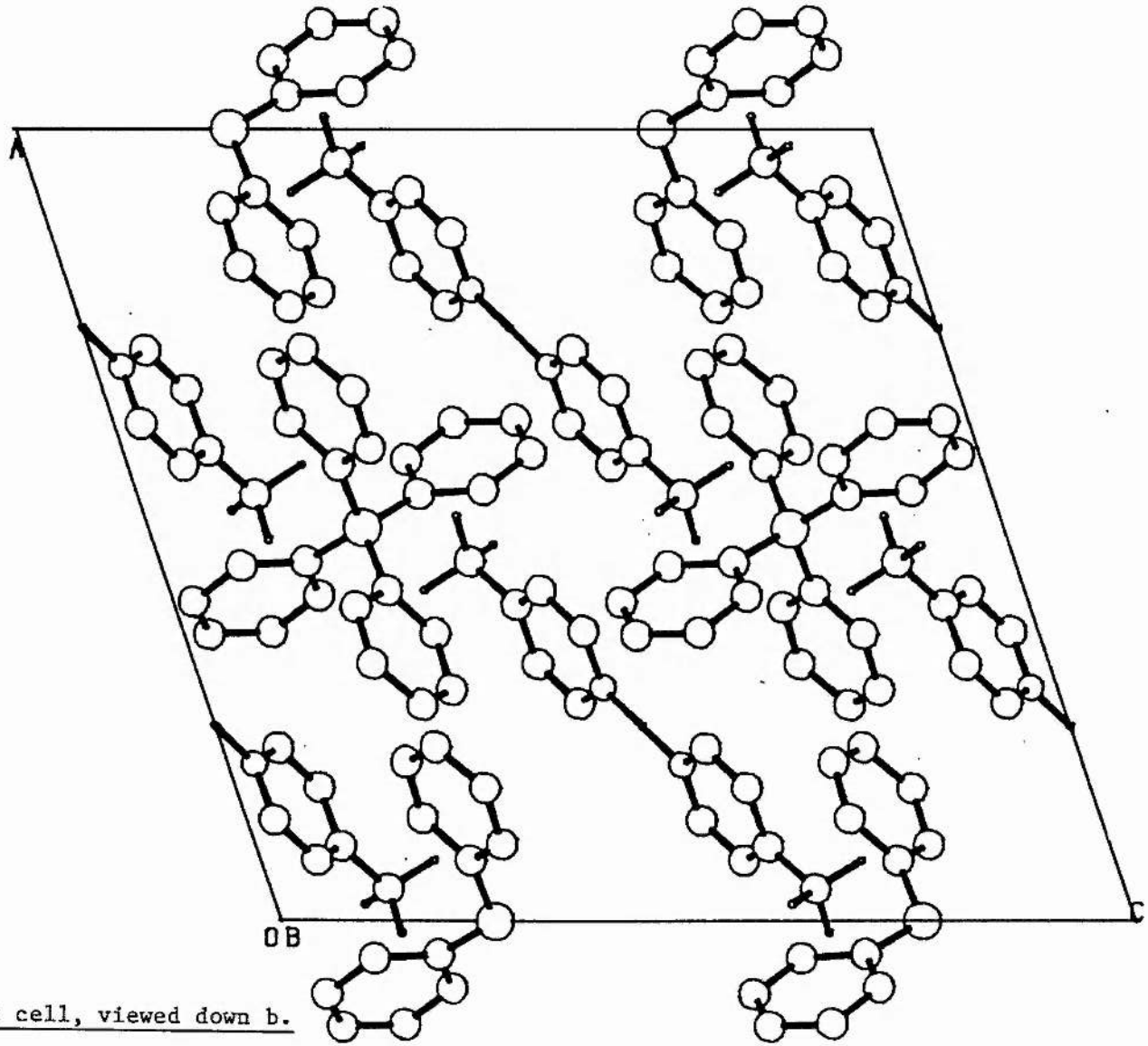

Figure 4.1.2. The unit cell, viewed down b.

Figure 4.1.1. Bis(4-methylpyridine)hydrogen(I) tetraphenylborate.

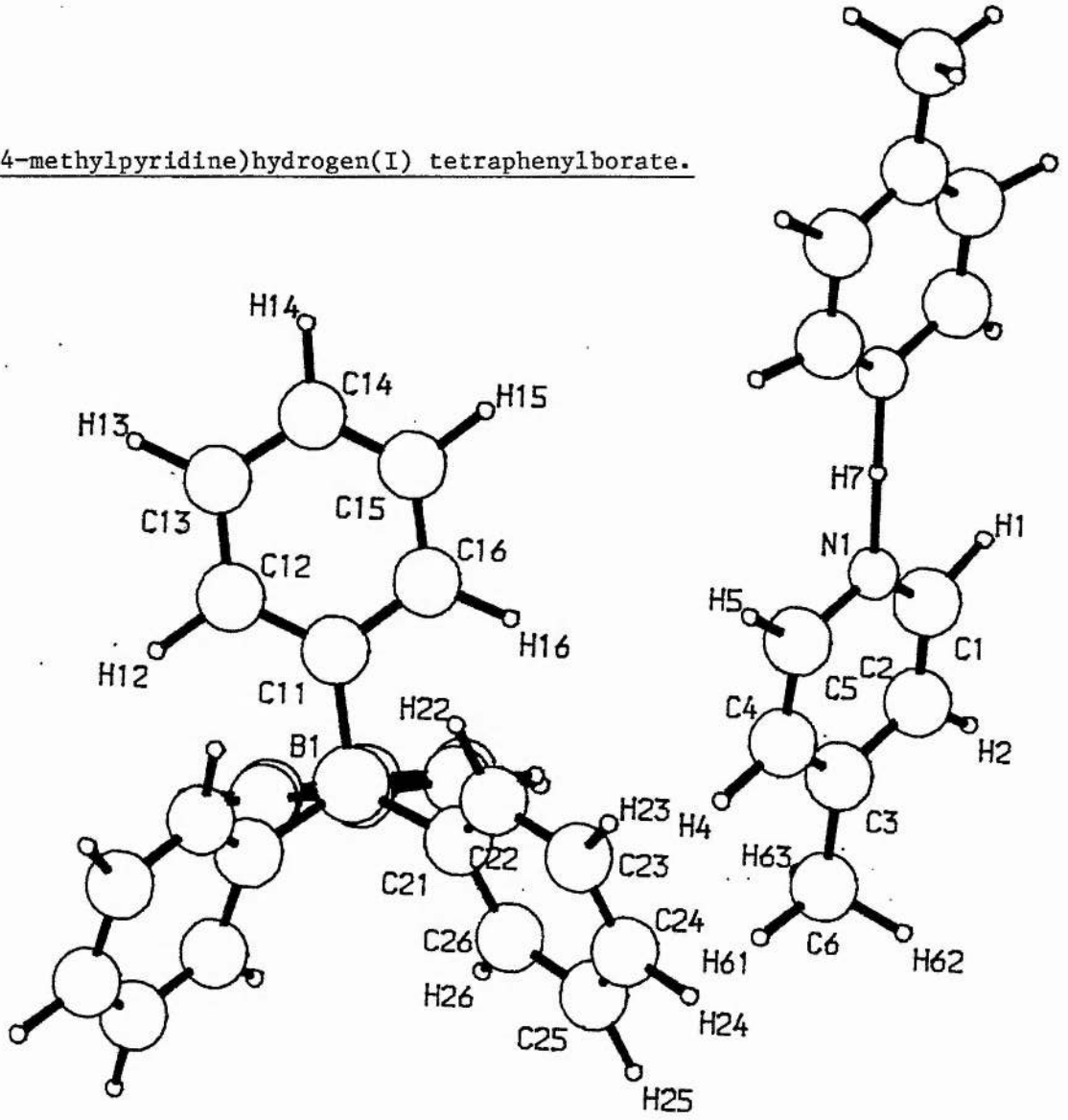

Table 4.1.1. Fractional Atomic Coordinates (x 10^4; for H x 10^3).

Atom	x	y	z
B(1)	5000(0)	4839(11)	7500(0)
C(11)	4213(3)	3953(7)	7542(3)
C(12)	4007(4)	2829(9)	7113(5)
C(13)	3276(5)	2124(10)	7084(5)
C(14)	2767(5)	2575(12)	7506(6)
C(15)	2953(4)	3662(11)	7905(5)
C(16)	3665(4)	4374(8)	7938(4)
C(21)	5426(3)	5689(7)	8291(3)
C(22)	5482(3)	5248(7)	9064(4)
C(23)	5935(4)	5918(9)	9762(4)
C(24)	6352(4)	6974(10)	9707(5)
C(25)	6323(4)	7469(8)	8975(5)
C(26)	5852(4)	6796(8)	8274(4)
N(1)	2984(4)	8211(8)	9688(4)
C(1)	2780(5)	9302(9)	9347(5)
C(2)	3278(5)	9972(9)	9020(4)
C(3)	4031(5)	9391(9)	9005(5)
C(4)	4218(5)	8252(11)	9350(6)
C(5)	3697(7)	7638(10)	9674(6)
C(6)	4586(5)	84(12)	8624(6)
H(12)	429(4)	231(7)	672(4)
H(13)	323(4)	125(7)	677(4)
H(14)	225(4)	209(6)	763(4)
H(15)	265(4)	414(7)	832(4)
H(16)	378(4)	518(7)	816(4)
H(22)	523(4)	448(7)	910(4)

H(23)	592(4)	543(6)	1032(4)
H(24)	658(4)	749(6)	1020(4)
H(25)	646(4)	837(7)	886(4)
H(26)	589(4)	730(7)	779(4)
H(1)	234(5)	993(7)	944(4)
H(2)	310(4)	1097(8)	878(4)
H(4)	467(5)	764(7)	918(4)
H(5)	380(5)	683(8)	994(5)
H(7)	250(0)	-250(0)	1000(0)

Table 4.1.2. Geometry of the Cation.

(a) Distances(Å).

N(1) ... N(1)'	2.610(15)	N(1) - H(7)	1.305(7)
N(1) - C(1)	1.275(12)	C(1) - H(1)	1.01(8)
N(1) - C(5)	1.301(13)	C(2) - H(2)	1.12(8)
C(1) - C(2)	1.312(13)	C(4) - H(4)	1.08(9)
C(2) - C(3)	1.363(12)	C(5) - H(5)	0.95(8)
C(3) - C(4)	1.319(15)		
C(4) - C(5)	1.307(16)		
C(3) - C(6)	1.452(14)		

(b) Angles(°).

C(1) - N(1) - C(5)	120.5(8)	C(2) - C(3) - C(4)	118.4(9)
C(1) - N(1) - H(7)	126.4(7)	C(2) - C(3) - C(6)	117.4(9)
C(5) - N(1) - H(7)	112.8(8)	C(4) - C(3) - C(6)	124.2(9)
N(1) - C(1) - C(2)	124.4(8)	C(3) - C(4) - C(5)	122.9(9)
C(1) - C(2) - C(3)	115.7(9)	C(4) - C(5) - N(1)	117.8(10)

Table 4.1.3. Geometry of the Anion.

(a) Distances(Å).

B(1) - C(11)	1.591(9)	B(1) - C(21)	1.560(9)
C(11) - C(12)	1.365(11)	C(21) - C(22)	1.351(9)
C(11) - C(16)	1.339(10)	C(21) - C(26)	1.354(10)
C(12) - C(13)	1.377(12)	C(22) - C(23)	1.358(9)
C(13) - C(14)	1.330(15)	C(23) - C(24)	1.313(13)
C(14) - C(15)	1.307(16)	C(24) - C(25)	1.320(12)
C(15) - C(16)	1.354(12)	C(25) - C(26)	1.370(10)
C(12) - H(12)	1.06(8)	C(22) - H(22)	0.91(7)
C(13) - H(13)	1.04(7)	C(23) - H(23)	1.08(7)
C(14) - H(14)	1.04(7)	C(24) - H(24)	0.96(7)
C(15) - H(15)	1.09(8)	C(25) - H(25)	1.00(7)
C(16) - H(16)	0.92(7)	C(26) - H(26)	0.99(7)

(b) Angles(°).

C(11) - B(1) - C(21)	115.7(3)	C(11) - B(1) - C(21´)	103.6(4)
C(11) - B(1) - C(11´)	108.5(3)	C(21) - B(1) - C(21´)	110.3(3)
B(1) - C(11) - C(12)	122.9(6)	B(1) - C(21) - C(22)	119.6(6)
B(1) - C(11) - C(16)	119.4(7)	B(1) - C(21) - C(26)	124.0(6)
C(16) - C(11) - C(12)	117.4(6)	C(26) - C(21) - C(22)	115.9(6)
C(11) - C(12) - C(13)	122.8(8)	C(21) - C(22) - C(23)	120.0(7)
C(12) - C(13) - C(14)	117.2(9)	C(22) - C(23) - C(24)	121.3(7)
C(13) - C(14) - C(15)	120.2(9)	C(23) - C(24) - C(25)	122.2(7)
C(14) - C(15) - C(16)	123.6(9)	C(24) - C(25) - C(26)	115.9(9)
C(15) - C(16) - C(11)	118.8(8)	C(25) - C(26) - C(21)	124.6(7)

II. Bis(triphenylarsineoxide)hydrogen(I) tetrafluoroborate.

Preparation.

Suitable crystals for X-ray work were kindly given by Dr. G.S. Harris and Mr. J.S. McKechnie, having been prepared by reaction of triphenylarsine oxide with a 1% solution of hydrogen fluoride in a glass vessel over a period of 24 hours[5]. This compound was originally thought to have the molecular formula $C_{36}H_{34}As_2F_4O_2$ but was later shown to be $C_{36}H_{31}As_2BF_4O_2$.

Crystal Data.

$C_{36}H_{31}As_2BF_4O_2$ is monoclinic, space-group $P2_1/c$ (C_{2h}^5, No.14). a = 12.926(4), b = 17.819(6), c = 14.994(4) Å, β = 98.87(3)°, U = 3412.2 Å^3. M_r = 732.29, D_c = 1.410 g.cm.$^{-3}$ for Z = 4. F(000) = 1472.00. Graphite monochromatised Mo-K_α radiation used with λ = 0.71069 Å, μ(Mo-K_α) = 19.32 cm^{-1}.

Data Collection and Structure Solution.

The intensities of 4894 unique reflections having $2^\circ \leqslant \theta \leqslant 30^\circ$ were measured in the quadrant $\pm h$, $+k$, $+l$, for k = 0 - 19 ($0^\circ \leqslant \mu \leqslant 22.265^\circ$) using a Stoe STADI-2 two circle diffractometer in the ω-2θ scan mode. Each scan comprised 120 steps, each of 0.01° in

ω, measured for 0.6s. per step. Backgrounds were measured at each end of the peak scan for a time of (scan time/2) and standard reflections, which were measured after every fifty reflections showed only small random deviations about their means. Then, Lorentz and polarisation corrections were applied to the data, but no absorption corrections made. 2497 unique reflections with $F_o \geqslant 6\sigma(F_o)$ were used for structure solution and refinement.

Systematic absences for space-group $P2_1/c$ are: h01: $1 \neq 2n$; 0k0: $k \neq 2n$. The structure was solved using direct methods with SHELX-76[2]. 326 reflections with E>1.6 gave 5999 triple-phase relations, used to calculate a convergence map. Phases -2 5 12, 1 2 11, 4 2 11, were used to fix the origin; and phases -8 1 12, -6 1 8, -2 1 12 and -6 5 8 for the multisolution tangent refinement to give 32 starting sets. The best E-map yielded suitable positions for one arsenic and ten carbon atoms.

Refinement continued satisfactorily showing the presence of the bis(triphenylarsineoxide)hydrogen(I) cation. When all non-hydrogen atoms of the cation had been located, and with the two arsenic atoms anisotropic, R was 0.1672 and Rg 0.2032, the only other electron density of significance in the difference map was a tetrahedral cluster of 5 peaks.

At first, it was thought that the stoichiometry of this compound corresponded to an n:2n addition of the reactants viz. $nPh_3AsO.2nHF$. Since the cation was known to be present from X-ray and infra-red data, which showed no evidence for the $(Ph_3AsOH)^+$ cation (no O-H stretch in the region 3000-2800 $cm.^{-1}$ normally found in the spectra of such compounds) but did indeed have a strong broad absorption in the range 1600-600 $cm.^{-1}$ (please see Figure 5.4. in following chapter), indicative of the O-H stretching mode in a strongly hydrogen bonded ion, it seemed reasonable to postulate that the compound under study had the formula $C_{36}H_{34}As_2F_4O_2$, and hence the anion present to be $H_3F_4^-$. At this stage, this postulate was amply supported by the results of microanalysis for 4 elements $(C,H,^{(\neq)}F,As^{(\neq)})$ which were in very reasonable accord with stoichiometry $C_{36}H_{34}As_2F_4O_2$.

Giving all 5 peaks of this tetrahedral cluster complex neutral-atom scattering factors[3,4] for fluorine, but refining the site occupation factors demonstrated that only the outer electron density peaks of this tetrahedron were genuinely fluorines. A tetrahedral arrangement of fluorine atoms with a proton in the middle and two disordered protons lying outside the tetrahedron proper was deemed to be possible (but with reservation) on the basis that:

(i) Although the fluorine atoms always had very large thermal parameters, the isotropic thermal parameter for the proton in the centre of the tetrahedron tended to refine to the very small value of 0.001, thus indicating that the

wrong type of atom was thought to be there.

(ii) Rigid body analysis[8,9] for the anion, which takes into account libration of the fluorine atoms and hence corrects bonding and non-bonding distances was unsuccessful.

Simultaneously, ^{1}H and ^{9}F n.m.r. spectra indicated that there were no proton resonances which could possibly be attributable to the protons of this anion. Also there was only one peak observable in the ^{9}F n.m.r. spectrum (whereas surely in $H_3F_4^-$ splitting of the ^{9}F resonance would be expected) occurring at exactly the correct frequency for BF_4^-! Then microanalysis for boron was carried out[(≠)] and the microanalysis results were excellent for molecular formula $C_{36}H_{31}As_2BF_4O_2$. The final cycles of blocked full-matrix least-squares refinement were carried out for the cation and the BF_4^- anion: R converged at 0.1060 and Rg at 0.1187 for all non-hydrogen atoms anisotropic and for this anion rigid body analysis[8,9] is wholly successful (please see Table 4.2.3). 407 parameters were varied, comprising 135 positional coordinates, 270 anisotropic thermal parameters, 1 common isotropic thermal parameter for the phenyl hydrogens and an overall scale factor. In this case neither cation nor anion lie on any special symmetry element, and no electron density peak could be located for the unique hydrogen between O(1) and O(2) in the cation, although the O...O distance is very short, 2.436(21) Å.

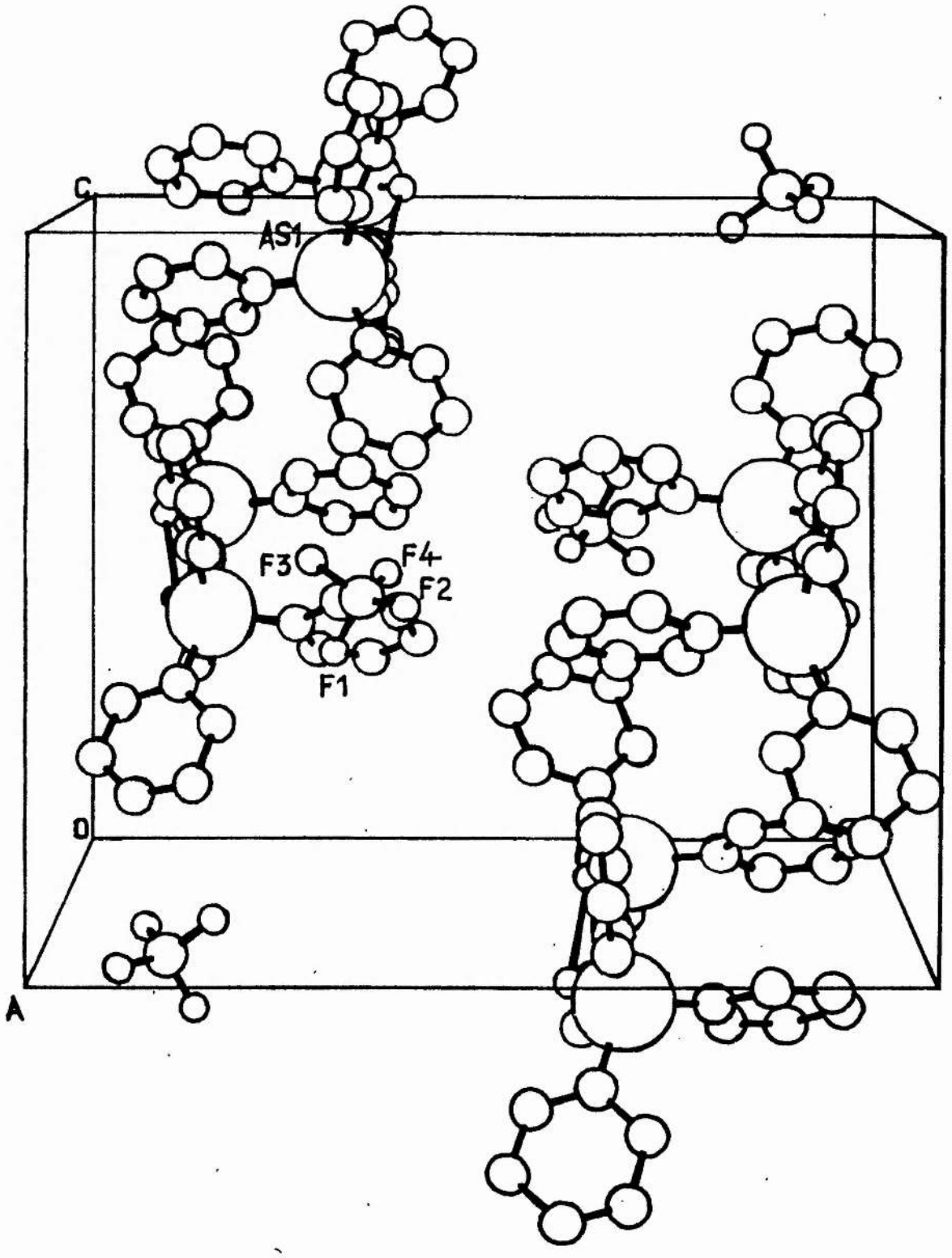

Figure 4.2.2. The contents of the unit cell, viewed down a.

Figure 4.2.1. Bis(triphenylarsineoxide)hydrogen(I) tetrafluoroborate.

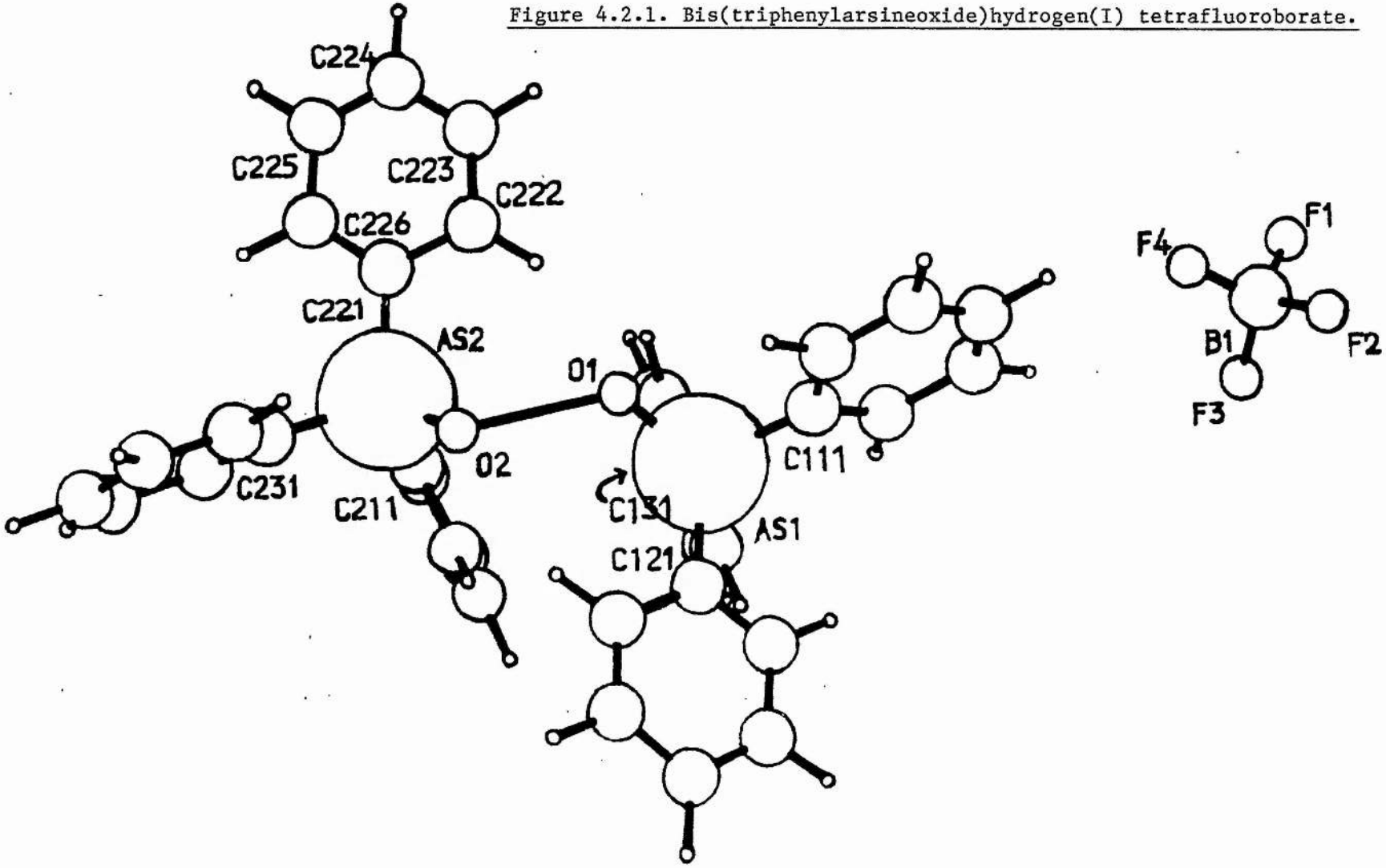

Table 4.2.1. Fractional Atomic Coordinates (x 10^4; for H x 10^3).

Atom	x	y	z
F(1)	6626(28)	3323(17)	3938(13)
F(2)	7610(16)	4144(14)	4711(19)
F(3)	7233(24)	3067(15)	5303(15)
F(4)	6052(19)	3921(21)	4955(20)
B(1)	6883(22)	3641(20)	4733(20)
As(1)	3204(2)	3296(1)	8991(1)
O(1)	2087(11)	3774(8)	8971(9)
C(111)	3840(15)	3662(11)	8040(12)
C(112)	4409(18)	3171(13)	7547(12)
C(113)	4867(19)	3433(17)	6759(15)
C(114)	4693(20)	4153(16)	6537(16)
C(115)	4135(20)	4654(14)	6989(15)
C(116)	3705(19)	4376(15)	7728(13)
C(121)	4152(16)	3464(12)	83(13)
C(122)	3777(17)	3730(13)	837(13)
C(123)	4481(25)	3823(16)	1618(15)
C(124)	5541(23)	3608(16)	1718(16)
C(125)	5861(16)	3371(14)	957(16)
C(126)	5180(19)	3276(14)	147(18)
C(131)	2918(18)	2252(14)	8792(15)
C(132)	2041(22)	2017(20)	8273(20)
C(133)	1752(25)	1356(22)	8104(24)
C(134)	2447(28)	771(20)	8494(21)
C(135)	3379(24)	974(15)	9043(18)
C(136)	3638(18)	1706(15)	9212(14)

As(2)	27(2)	3436(1)	180(1)
O(2)	1066(11)	4002(8)	181(9)
C(211)	503(17)	2421(13)	338(14)
C(212)	-182(19)	1831(15)	9960(16)
C(213)	217(29)	1145(19)	100(26)
C(214)	1205(27)	977(19)	552(18)
C(215)	1831(23)	1561(19)	920(19)
C(216)	1455(20)	2252(18)	798(18)
C(221)	-965(15)	3570(14)	9097(14)
C(222)	-557(19)	3606(15)	8263(17)
C(223)	-1284(27)	3738(15)	7492(16)
C(224)	-2328(28)	3747(21)	7496(18)
C(225)	-2693(21)	3680(27)	8282(20)
C(226)	-2001(21)	3602(22)	9098(20)
C(231)	-591(16)	3745(13)	178(14)
C(232)	-1054(19)	3205(12)	1674(13)
C(233)	-1515(20)	3414(14)	2464(13)
C(234)	-1490(19)	4121(16)	2688(17)
C(235)	-1057(23)	4702(16)	2184(20)
C(236)	-629(22)	4454(16)	1442(17)
H(112)	451(2)	259(1)	776(1)
H(113)	532(2)	307(2)	639(2)
H(114)	501(2)	436(2)	596(2)
H(115)	404(2)	523(1)	678(2)
H(116)	324(2)	475(2)	807(1)
H(122)	296(2)	386(1)	1082(1)
H(123)	420(3)	408(2)	1219(2)
H(124)	606(2)	363(2)	1236(2)

H(125)	668(2)	325(1)	1097(2)
H(126)	547(2)	305(1)	957(2)
H(132)	152(2)	245(2)	796(2)
H(133)	101(3)	123(2)	769(2)
H(134)	226(3)	19(2)	836(2)
H(135)	390(2)	54(2)	935(2)
H(136)	436(2)	187(2)	964(1)
H(212)	-96(2)	194(2)	960(2)
H(213)	-27(3)	68(2)	982(3)
H(214)	147(3)	40(2)	1063(2)
H(215)	260(2)	146(2)	1129(2)
H(216)	194(2)	271(2)	1109(2)
H(222)	27(2)	354(2)	823(2)
H(223)	-100(3)	384(2)	686(2)
H(224)	-286(3)	381(2)	687(2)
H(225)	-353(2)	368(3)	829(2)
H(226)	-231(2)	357(2)	973(2)
H(232)	-107(2)	262(1)	1147(1)
H(233)	-187(2)	300(1)	1285(1)
H(234)	-181(2)	428(2)	1328(2)
H(235)	-107(2)	529(2)	1237(2)
H(236)	-30(2)	488(2)	1105(2)

Table 4.2.2. Geometry of $C_3AsO....OAsC_3$.

(a) Distances (Å).

O(1) ... O(2)	2.436(21)		
As(1) - O(1)	1.672(15)	As(2) - O(2)	1.679(15)

As(1) - C(111)	1.869(20)	As(2) - C(211)	1.913(22)
As(1) - C(121)	1.910(19)	As(2) - C(221)	1.923(19)
As(1) - C(131)	1.911(24)	As(2) - C(231)	1.884(22)

(b) Angles(°).

O(1) - As(1) - C(111)	106.8(8)	O(2) - As(2) - C(211)	109.0(8)
O(1) - As(1) - C(121)	111.9(8)	O(2) - As(2) - C(221)	110.8(8)
O(1) - As(1) - C(131)	110.3(8)	O(2) - As(2) - C(231)	105.1(8)
C(111)- As(1) - C(121)	107.5(9)	C(211)- As(2) - C(221)	112.5(9)
C(111)- As(1) - C(131)	108.5(9)	C(211)- As(2) - C(231)	110.2(9)
C(121)- As(1) - C(131)	111.7(9)	C(221)- As(2) - C(231)	109.0(9)
As(1) - O(1) - O(2) - As(2)	99.5(2)		

Table 4.2.3. Geometry of the BF_4^- Anion.

(a) Distances (Å).

	uncorrected	corrected for libration	
B(1) - F(1)	1.314(38)	1.471	
B(1) - F(2)	1.302(40)	1.434	
B(1) - F(3)	1.364(41)	1.500	mean 1.461
B(1) - F(4)	1.274(42)	1.439	
F(1)...F(2)	2.155(38)	2.417	
F(1)...F(3)	2.125(31)	2.331	
F(1)...F(4)	2.088(43)	2.346	
F(2)...F(3)	2.199(38)	2.394	mean 2.383
F(2)...F(4)	2.138(35)	2.371	
F(3)...F(4)	2.161(42)	2.411	

(b) Angles(°).

F(2) - F(1) - F(3)	61.8(12)	F(1) - F(4) - F(2)	61.3(14)
F(2) - F(1) - F(4)	60.5(12)	F(1) - F(4) - F(3)	60.0(13)
F(3) - F(1) - F(4)	61.7(13)	F(2) - F(4) - F(3)	61.5(13)
F(1) - F(2) - F(3)	58.4(11)	F(1) - B(1) - F(2)	110.9(30)
F(1) - F(2) - F(4)	58.2(14)	F(1) - B(1) - F(3)	105.0(29)
F(3) - F(2) - F(4)	59.8(14)	F(1) - B(1) - F(4)	107.5(28)
F(1) - F(3) - F(2)	59.8(13)	F(2) - B(1) - F(3)	111.1(25)
F(1) - F(3) - F(4)	58.3(14)	F(2) - B(1) - F(4)	112.1(32)
F(2) - F(3) - F(4)	58.7(12)	F(3) - B(1) - F(4)	110.0(30)

Chapter Four.

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≠ Microanalysis carried out by S.Smith, Chemistry Department, St. Andrews (C,H).

≠ Microanalysis carried out by A.Bernhardt, Max Planck Institut, Mulheim, Germany (As,F,B).

Chapter Five.

Some considerations following the X-ray work of Chapter Four.

I. Introduction.

Although these compounds are very different, one being an organometallic, the other a heterocyclic compound, their common feature is that both contain a bis cation which has very strong hydrogen bonding as shown by the short O(1)...O(2) and N(1)...N(1)' distances of 2.436(21) Å and 2.610(15) Å respectively.

II. The (Amine-H-Amine)$^+$ cation.

The salient feature, which prompted the X-ray structure determination of bis(4-methylpyridine)hydrogen(I) tetraphenylborate, was investigation of whether or not this bis cation did indeed exist in the solid state. Its existence in some lanthanoid chelates and tetraphenylborates had been previously suspected by Glidewell and Shepherd in 1975[1] but not proven.

Firstly, a series of compounds with stoichiometry $Ln(hfaa)_4.L.LH$ where Ln is a Lanthanoid element, hfaa = $(CF_3COCHCOCF_3)^-$ and L = 3- or 4-methylpyridine had been prepared. Three suggestions may be made concerning the fate of the seemingly extra neutral amine molecule:

(i) The neutral molecule may coordinate to the lanthanoid. This would involve a nine-coordinate stereochemistry which is not unknown, but is more common when small monodentate ligands are chelated eg. $[Ln(H_2O)_9]^{3+}$.

(ii) The neutral molecule may only be present as solvent of crystallisation in the solid state.

(iii) Instead of having the simple pyridinium cation, $[LH]^+$, the neutral pyridine may complex to the cation giving the bis cation, $[L_2H]^+$.

It seemed likely that the neutral molecule was not involved in the lanthanoid coordination sphere, because another series, containing the large tetraphenylborate anion and having analogous stoichiometries $BPh_4.L.LH$ can also be prepared. Coordination of L is not possible in these compounds, and so (i) becomes a less viable proposition. So, also, does (ii) because the neutral molecule is not easily lost when the compounds are vacuum sublimed.

X-ray photoelectron spectroscopy (ESCA) on 4-methylpyridine and related compounds proved inconclusive in indicating whether (4-methylpyridine)H^+ or (4-methylpyridine)$_2H^+$ was present[(2)].

Compound	N(1s) Binding Energy (eV)
4-mepy	398.1
4-mepy.HCl	400.2
4-mepyH.BPh_4.4-mepy	399.6
4-mepyH.BPh_4.CH_3CN	400.7, 399.5

Whilst the spectrum of 4-mepyH.BPh_4.4-mepy gave one band (indicating equivalence of nitrogens and the bis cation) because of a shoulder in the band at lower binding energy, the band could be a merged doublet. In which case, the simple cation and solvent of crystallisation would be present.

Two N(1s) bands are present in the spectrum of 4-mepyH.BPh_4CH_3CN. Since these cannot be specifically assigned to either of the nitrogens present, possible presence of either the monocation or the complex (4-methylpyridine)(acetonitrile)hydrogen(I) cation remains.

So, existence of the bis cation was postulated on the basis of such work, but an X-ray structure was necessary to provide unequivocal evidence.

III. Existence of analogous (Amine-Halogen-Amine)$^+$ species.

Similar cationic complexes have also been reported with heavier halogens as the central atom[3]. X-ray structures for two iodine complexes have been reported in the literature. Hassel and Hope, working with bis(pyridine)iodine(I) heptaiodide in 1961[4] showed that centrosymmetric py_2I^+ cations were present (N - I distance of 2.16(10) Å) which were almost planar. The remaining iodine atoms form I_3^- anions, linked by neutral I_2 molecules, leading to an arrangement resembling the heptaiodide anion.

Pritzkow has investigated bis(hexamethylenetetramine)iodine(I) triiodide[5] in which the cation is not strictly centrosymmetric but angle N-I-N in the cation is almost linear, 176.5(4)°. Pritzkow states that the N-I distance of 2.30(1) Å is slightly larger than that found in the bis(3-methylpyridine)iodine(I) cation (2.24(2) Å) by Osborn.

Data for only one bromine complex exists[6], in bis(quinoline)bromine(I) perchlorate. There is <u>almost</u> a centre of symmetry at Br, the quinoline groups being coplanar in trans configurations. The N-Br-N arrangement is linear (average N-Br-N angle 177.0(4)°) with a mean N-Br distance of 2.13 Å but with each cation containing a long and short N-Br bond.

Stability of complex increases with Z, the halogen atomic number. Aromatic amine complexes of chlorine are known[2,7], mainly as the nitrate salts[8], but none have been studied crystallographically: the possibility remains of these not being true bis cations. Since fluorine is such an electronegative element it is difficult to envisage its ever forming complexes with neutral electron donors in a cationic complex readily. Solid $(quinoline)_2Br^+$ appears to be not particularly stable, because decomposition is mentioned and several different crystals were needed[6]. Slight decomposition is also mentioned by Pritzkow for bis(hexamethylenetetramine)iodine(I) triiodide[5].

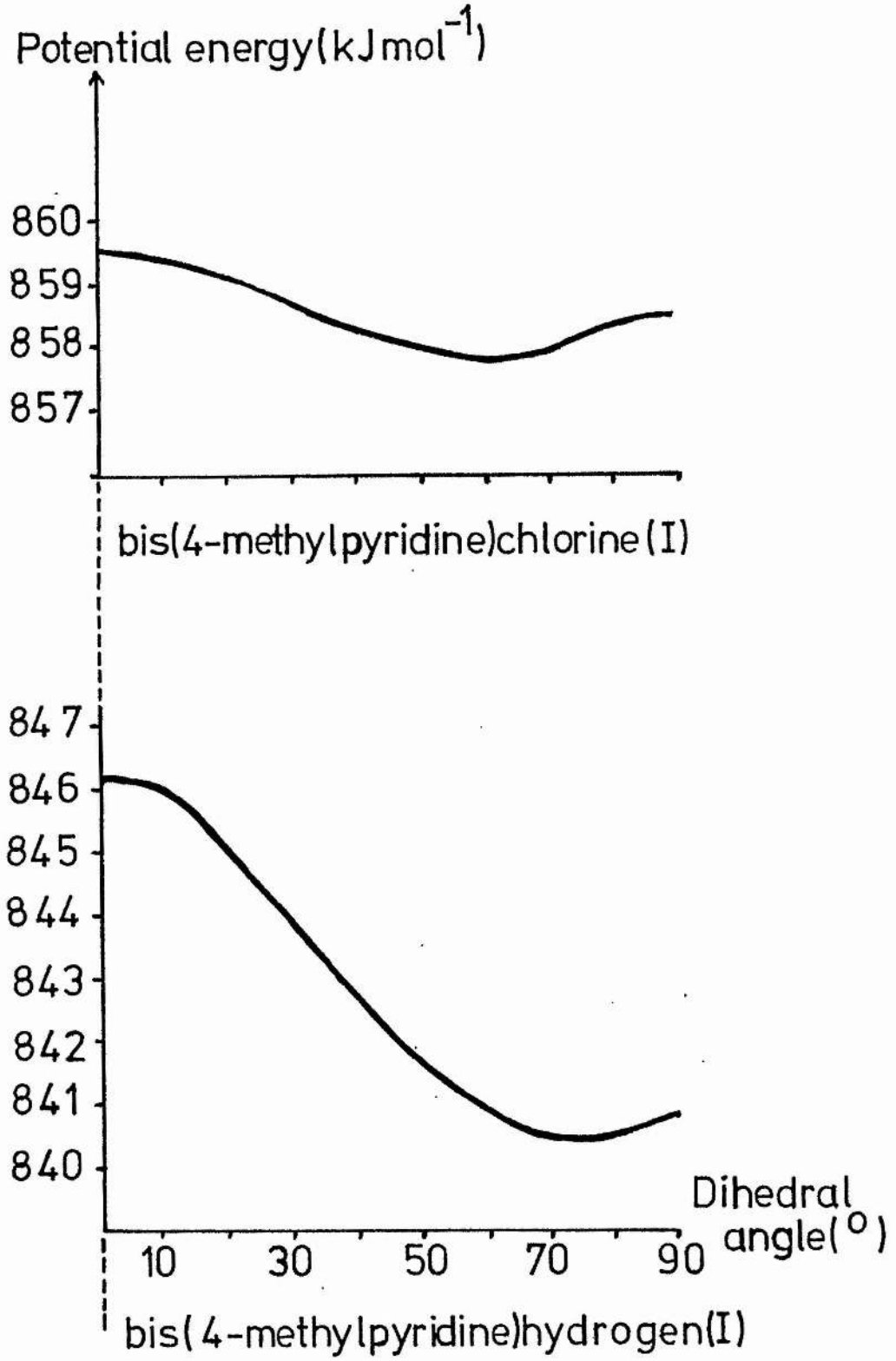
IV. Planar or Non-planar Complexes?

In the structure determination reported in the previous chapter, the bis(4-methylpyridine)hydrogen(I) cation lies across a centre of inversion, and hence the cation is crystallographically required to be planar. If, however, free rotation of the 4-methylpyridine groups is allowed, when the optimum geometry for this cation is calculated (in the gas phase, using the MNDO[9] programme system) then the preferred geometry is non-planar, with a dihedral angle of 75.3° between the two rings. Bis(pyridine)iodine(I) in the heptaiodide is also nearly planar, and centrosymmetric[4]. Bis(quinoline)bromine(I) perchlorate has an almost linear, although asymmetric N-Br-N skeleton with the two quinolines almost coplanar and trans to one another. The cation therefore has approximate centrosymmetry[6].

Bis(thiourea)iodine(I) iodide[10] has also been investigated and crystallises in space-group C2/c, paralleling our work. It, too, has a centrosymmetric cation, centre of inversion 1/4, 1/4, 0 with the anion, I^-, lying on a two-fold axis at 0, y, 1/4. The S-I-S arrangement is therefore linear, the ligands being non-planar.

MNDO calculations have been carried out for the bis(4-methylpyridine)hydrogen(I) cation in an attempt to find the energy barrier to rotation of one ring with respect to the other, when this cation is considered as an isolated species in the gas phase. This was done by taking the geometry of the optimised species and fixing the dihedral angle between the two rings at certain values from 0^o, 10^o, 20^o ....90^o, and then performing one SCF calculation. The calculated heat of formation is taken as a measure of the potential energy of the species. The results are summarised in Table 5.1 and plotted in Figure 5.1, and show that the energy barrier to rotation for the isolated species is small, only ca. 6 $kJmol^{-1}$. This might be expected since only the 1s orbital of the proton is capable of gaining electrons and hence forming the 3 centre, 4 electron bonding scheme in the complex. Because the 1s orbital of the proton is of ^1A_g symmetry, then there are no geometric restrictions on orbital overlap of the lone pair of electrons being donated from the nitrogen of each 4-methylpyridine molecule, and so twisting of the complex will involve only very small changes of potential energy. In the crystal, efficient packing of this cation and counterion in order to lower the potential energy by ion-ion electrostatic interactions will be the

Figure 5.1. Variation of Cation Potential Energy with Dihedral Angle between rings.



dominating factor. Thus the cation may twist from the optimum gas phase geometry to fit more snugly into the crystalline lattice.

Table 5.1.

Hydrogen(I) complex		Chlorine(I) complex	
Dihedral angle (^{o})	Heat of formation ($kJmol^{-1}$)	Dihedral angle (^{o})	Heat of formation ($kJmol^{-1}$)
0	846.329	0	859.520
10	846.021	10	859.418
20	845.179	20	859.139
30	844.024	30	858.752
40	842.804	40	858.344
50	841.721	50	858.000
60	840.926	60	857.808
70	840.560	70	857.880
80	840.654	80	858.240
90	840.813	90	858.490

The barrier to rotation for the bis(4-methylpyridine)chlorine(I) complex is extremely small when calculated in a similar fashion. Admittedly, only s and p orbitals were involved in the MNDO calculations ($p_{\pi'}-d_{\pi'}$ interaction may make some difference, but this is unlikely since interaction will be the same at 0^{o} and 90^{o} because of orbital symmetry) however the rotation barrier is so small it will be negligible compared to intermolecular forces. The slight preference for the twisted hydrogen(I) cation may tentatively be

attributed to steric interaction of ring hydrogens.

The molecule therefore adopts the shape representing the most favourable balance of forces acting upon it.

V. Stabilities of some Amine Complexes.

Calculations were done, using the MNDO programme system[9], to find enthalpies of formation, in the gas phase, for several neutral amines; these as ligands to H^+, F^+, Cl^+ in simple cations; and then the corresponding bis cations. Unfortunately, MNDO calculations cannot be employed for the Br^+ and I^+ complexes since no atoms having d-electrons have been parameterised for the MNDO system. Nor could energies of some species with ligands complexed to F^+ be calculated: the species were so unstable that during SCF calculations, self-consistence could not be achieved. In the case of trimethylamine and $H_2NCH{=}CH_2$, the geometries of the bis cations could not be optimised, because one of the the N - H distances steadily increased.

The enthalpies of some known and hypothetical complexes, calculated by MNDO, are shown in Table 5.2.

Table 5.2. Heats of formation for Amine complexes.

Amine	Neutral L	$[LX]^+$		$[L_2X]^+$		$A^{(**)}$	$B^{(***)}$
NH_3	-26.33	(i)	689.21	(i)	644.48	662.88	-18.40
		(ii)	817.74	(ii)	774.12	791.41	-17.29

		(iii)	840.05	(iii)	666.73	813.72	-146.99
NF_3	-143.07	(i)	1018.76	(i)	943.93	875.69	68.24
		(ii)	1117.47	(ii)	no S C	974.40	
		(iii)	1166.98	(iii)	1009.94	1023.91	-13.97
pyridine	120.22	(i)	785.71	(i)	912.34	905.93	6.41
		(ii)	no S C				
		(iii)	917.14	(iii)	928.45	1037.36	-108.91
4-methyl pyridine	86.92	(i)	746.95	(i)	840.51	833.87	6.64
		(ii)	no S C				
		(iii)	877.54	(iii)	too many e^-'s		
NMe_3	-11.64	(i)	728.48	(i)	unstable		
		(ii)	857.19				
		(iii)	892.95				
$H_2NCH{=}CH_2$	58.66	(i)	769.38	(i)	unstable		

(*) All values quoted are in $kJmol^{-1}$.

(**) Column A gives the sum of the formation energy for $[LX]^+$ and energy of neutral L.

(***) Column B gives the difference between the formation energy for $[L_2X]^+$ and values of column A. Thus, these are a measure of the stabilities of $[L_2X]^+$ compared to $[LX]^+$ and L, in the gas phase.

(i),(ii),(iii). Lines marked thus, refer to the H^+, F^+ and Cl^+

complexes respectively.

Stability of the simple cation would be reflected in values of the gas phase pK_a's in the cases of the hydrogen(I) complexes. For the equilibrium

$$BH^+_{aq} + nH_2O \rightleftharpoons B_{aq} + H_3O^+_{aq}$$

where $K_a = \frac{(B)(H_3O^+)}{(BH^+)}$ and

$pK_a = -\log_{10}K_a$,

the larger the pK_a value, the less favoured is dissociation of BH^+. For example, some aqueous pK_a's taken from values given by Perrin are[11,12]:

Me_3N	>	H_3N	>>	4-mepy	≈	2-mepy	>	3-mepy	>	py	>	quinoline.
9.907		9.246		6.03		5.94		5.52		5.21		4.81

Trimethylamine and ammonia are very strong bases, as expected, because they very readily act as Lewis bases, donating their lone pair of valence electrons.

Of course, these pK_a values are for aqueous media and cations will be highly solvated. MNDO calculations are for isolated gas phase species, and appear to show the opposite trends insofar as the potential energy differences for forming the hydrogen(I) monocations are in the reverse order.

Table 5.3.

Amine	Neutral L	$[LH]^+$	Energy $[LH]^+ - L$
NH_3	-26.33	689.21	715.54
Me_3N	-11.64	728.48	740.12
pyridine	120.22	785.71	665.49
4-mepy	86.92	746.95	660.03

NB. The standard heat of formation of isolated H^+ is a constant throughout and $\Delta H_f^{\ominus} = 1530$ kJmol^{-1}.

Again, in Table 5.3, all energy values are quoted in kJmol^{-1}. The column 'Energy $[LH]^+ - L$' is a measure of the potential energy increase when the hydrogen(I) cation is formed, and the smaller this value, the more favoured will be cation formation in the gas phase.

The trend for this ease of formation is

$$NH_4^+ < Me_3NH^+ < pyridineH^+ < 4\text{-}mepyH^+$$

which as mentioned is the reverse order of aqueous solution pK_a values.

One might conclude that solvation effects in aqueous media will affect the stability of the small ammonium cation more than of large pyridine- or substituted pyridine- containing complexes.

Consider now column B of Table 5.2. A negative value in column B means that in the gas phase the bis cation is more stable than individual $[LX]^+ + L$. All bis(ligand)chlorine(I) complexes which are shown in Table 5.2 are more stable than their respective components. Bis(ammonia)hydrogen(I) and fluorine(I) are also more stable. Yet bis(pyridine)hydrogen(I) and bis(4-methylpyridine)hydrogen(I) are both less stable by ca. 6 $kJmol^{-1}$.

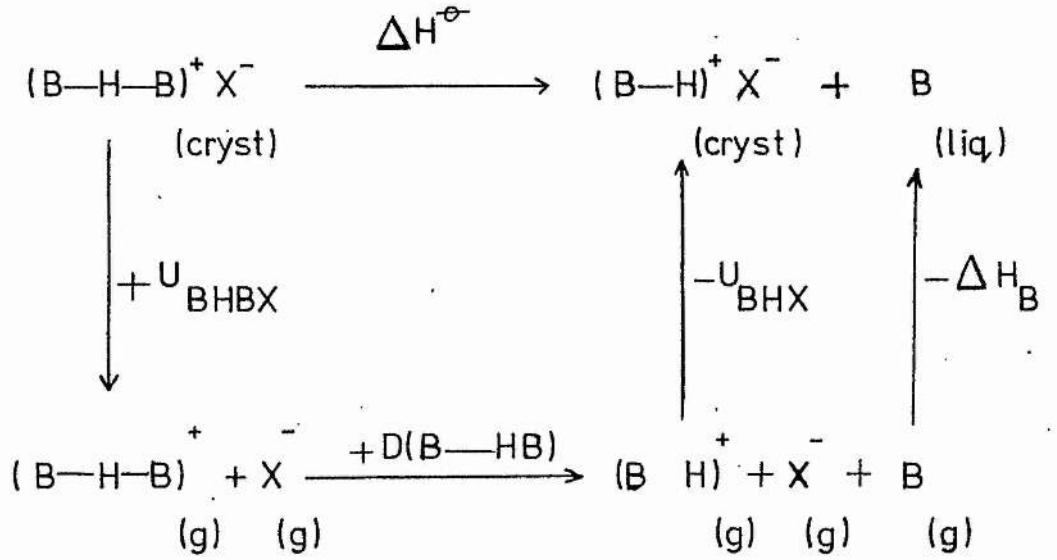
It must be stressed that these potential energies are for the gas phase, for isolated species. Solvation effects in aqueous solution alter the trends in stability, so is this also the case for the solid state? When these condense to form a solid crystalline lattice do electrostatic interactions lower the potential energy of the bis(pyridine)- and bis(4-methylpyridine)hydrogen(I) cations sufficiently to make them more stable? After all, 6 $kJmol^{-1}$ is not very much, and our crystallographic evidence shows this to be the case.

Why, then, should the bis(4-methylpyridine)hydrogen(I) cation be preferred to the simple cation plus neutral 4-methylpyridine of crystallisation? Greater delocalisation of charge may be an explanation, but the formal charge on the cation is only +1 overall,

and so one ring might be expected to delocalise this charge reasonably effectively.

The most likely explanation is stabilisation of a potentially unstable ion, (bis(4-methylpyridine)hydrogen(I)), by a large counter ion (BPh_4^-). This concept of stabilisation of an unstable ion has been widely used in the realm of organometallic chemistry where such bulky cations as the mu-nitrido-bis(triphenylphosphorus)(+1) (see also Chapter Six) and the tetraethylammonium cations have been widely used to stabilise a host of polynuclear carbonyl and other organometallic anions[13].

To explain this a Born-Haber cycle may be drawn[14]:



B is a base, eg. 4-methylpyridine. For this reaction, if B is a liquid or solid, then $\Delta S^{\ominus} \approx 0$ and therefore $\Delta H^{\ominus}$ approximates to $\Delta G^{\ominus}$.

$-U_{BHX}$, the lattice energy, and ΔH_B, the latent heat of evaporation will be exothermic: vapourisation of crystalline BHB^+X^-, ie. U_{BHBX} will be endothermic, as will D(B - HB), the bond dissociation energy.

From Kapustinskii's Equation (1) $\Delta U = U_{BHX} - U_{BHBX}$ will always be positive since $(BHB)^+$ is always the larger cation. For a given ligand, D and ΔH_B are fixed; therefore to stabilise the bis cation with respect to the monocation $\Delta H^{\ominus}$ must be increased and so ΔU minimised.

$$\Delta U = 2k \left[\frac{1}{r_{BH^+} + r_{X^-}} - \frac{1}{r_{BHB^+} + r_{X^-}} \right] \qquad (1)$$

(r represents the ionic radii)

$$\Delta U = 2k \left[\frac{r_{BHB^+} - r_{BH^+}}{(r_{BH^+} + r_{X^-})(r_{BHB^+} + r_{X^-})} \right]$$

Say $r_{BHB^+} = p.r_{BH^+}$: p is a constant, > 1

$$\therefore \Delta U = \frac{2k(p-1)\, r_{BH^+}}{(p.r_{BH^+} + r_{X^-})(r_{BH^+} + r_{X^-})} = \frac{2k(p-1)\, r_{BH^+}}{[pr^2_{BH^+} + (p+1).r_{BH^+}.r_{X^-} + r^2_{X^-}]}$$

Differentiating

$$\frac{\partial(\Delta U)}{\partial r_{BH^+}} = \frac{2k(p-1)(r_{X^-}^2 - p.r_{BH^+}^2)}{[p.r_{BH^+}^2 + (p+1)r_{BH^+}.r_{X^-} + r_{X^-}^2]^2} = 0$$

$$\text{if} \quad r_{X^-}^2 = p.r_{BH^+}^2$$

But since $p = \frac{r_{BHB^+}}{r_{BH^+}}$ and $\frac{\partial(\Delta U)}{\partial r_{BH^+}} = 0$, if $\frac{r_{BHB^+}}{r_{BH^+}} = \frac{r_{X^-}^2}{r_{BH^+}^2}$

that is, if $r_{BHB^+}.r_{BH^+} = r_{X^-}^2$, ΔU is minimised, $\Delta H^{\ominus}$ is increased and $(BHB)^+$ is stabilised. Therefore since $(BH)^+$ and $(BHB)^+$ are both comparatively large cations when B is 4-methylpyridine for example, X^- must also be large to have any effective stabilising ability. BPh_4^- is obviously sufficiently bulky.

Alcock mentions that crystals of bis(quinoline)bromine((I) perchlorate are rather unstable[6]. Had an anion larger than perchlorate been used, the crystals may have been more stable. Large counter ions render stability to large ions, and small counter ions stability to small ions.

VI. Reactions of Triphenylarsineoxide with aqueous hydrogen fluoride: Some other related compounds.

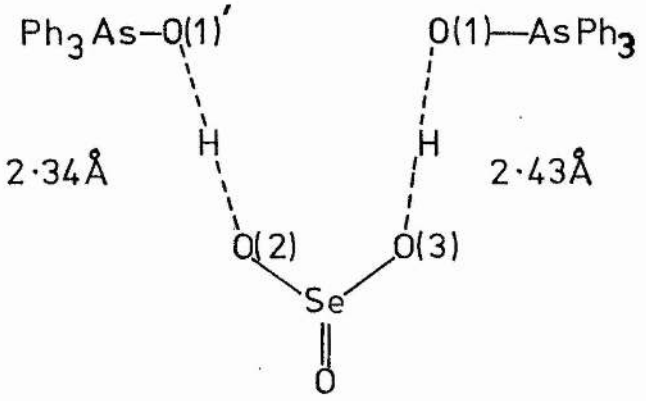
The preparation of bis(triphenylarsineoxide)hydrogen(I) tetrafluoroborate was carried out purely by chance. This was because the aqueous hydrogen fluoride - even at such high dilution, 1% - leached boron from the borosilicate glass during the relatively long reaction time (24 hours). Afterwards, various experiments were carried out by Mr. J.S. McKechnie using HF concentrations of 1 to 40% in plastic vessels. The product is triphenylarsinedifluoride. If excess 40% HF is added to triphenylarsineoxide in a glass vessel a precipitate of triphenylarsinedifluoride is obtained: reaction is too rapid for any boron-containing product to be formed[15].

The bis(triphenylarsineoxide)hydrogen(I) cation with $Hg_2Br_6^{2-}$ as counterion was previously reported[16], again with a short O...H...O distance 2.40(3) Å, and As - O 1.68(2) Å. One other example of the bis cation is reported in the crystallographic literature in 'triphenylarsineoxide semiperchlorate'[17]. The O-H...O hydrogen bond is 2.44 Å.

This bis cation is reported to hydrolyse to the $[Ph_3AsOH]^+$ cation in damp acetonitrile[15] which also has been reported by Harris *et al.*[18]. Infra-red studies of solids show strong OH absorption typically in the region 2800 - 3000 cm^{-1}. for this cation (cf. Figure 5.4).

The crystal structure of $Ph_3AsO.H_2O$ has been done by Ferguson[19]. The As - O bond length 1.644(7) Å is much shorter than in our bis cation (Although the hydrogens of the water molecule were not located, hydrogen bonding between the water and the oxygen of the triphenylarsineoxide group will be rather weak, the O....O distance being of the order 2.8 Å.

In contrast the 2:1 complex formed between triphenylarsineoxide and selenous acid is strongly hydrogen bonded[20].



The pyramidal SeO_3 group is disordered about a mirror plane. The As - O distance, 1.68(1) Å, is very similar to that in our cation, and is therefore considerably longer than in the monohydrate mentioned above. So, also, is there much stronger hydrogen bonding: O(1)....O(3), 2.43(3) Å; O(1)´....O(2), 2.34(3) Å.

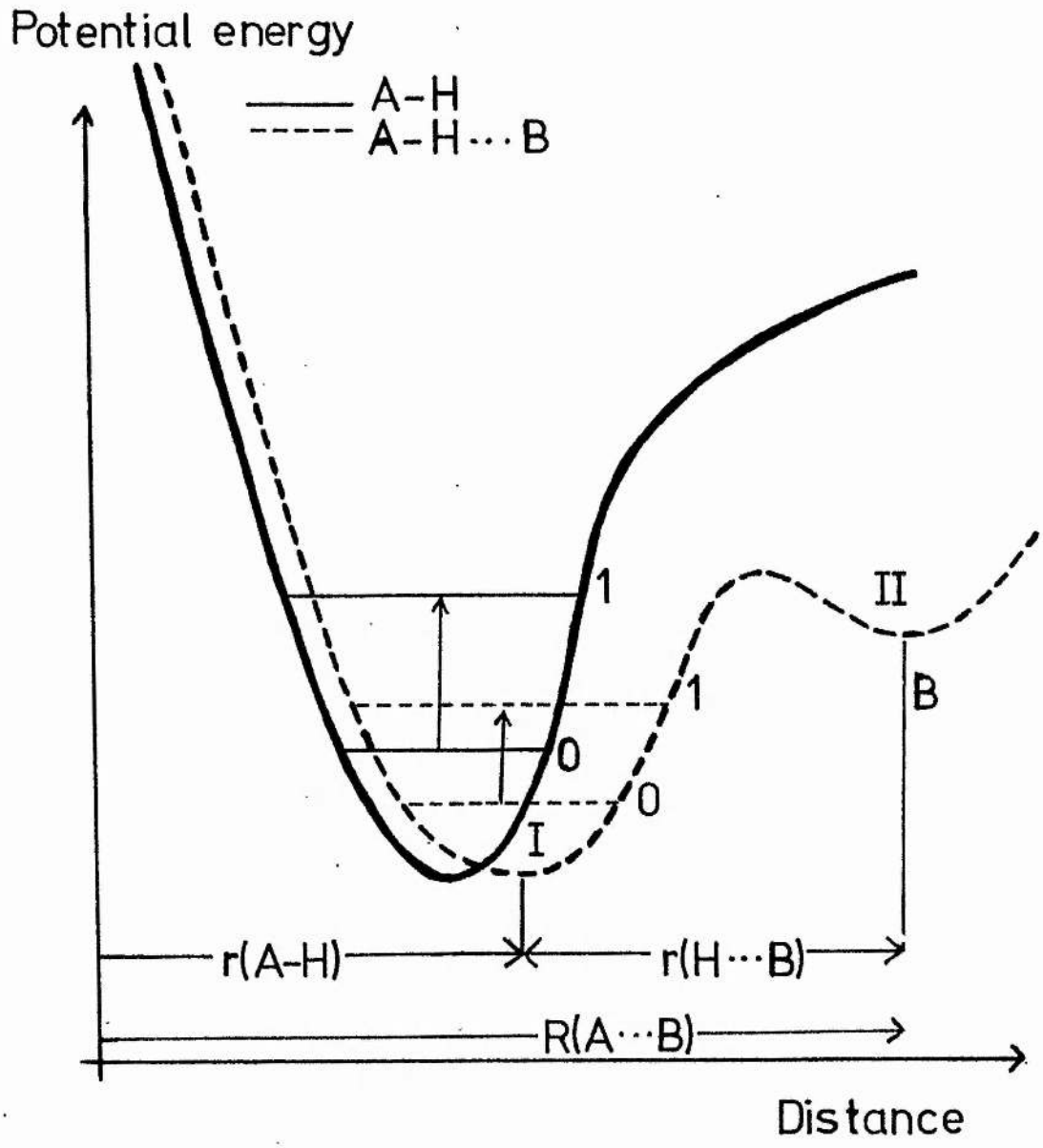
Obviously, if the As - O bonding is stronger (and shorter), less electron density will be available for the O - H...O hydrogen bonding: this is reflected in longer O....O distances.

VII. Strong (O...H...O) and (N...H...N) Hydrogen Bonding.

As mentioned previously, the common feature between these two compounds is both possess very strong hydrogen bonding in their cations. The O....O and N....N distances of 2.436(21) Å and 2.610(15) Å respectively fit in well with distances quoted by Novak[21] for strong hydrogen bonding in solids.

Hydrogen bonding manifests itself by a lowering of frequency and a broadening of the stretching bands in the infra-red and Raman spectra. This is because the potential energy function of, say, a free A - H group is modified when there is interaction in the form of hydrogen bonding, with another group B. The vibrational levels become closer together and so the A - H stretching frequency now occurs at a lower frequency and is further lowered and broadened as strength of the hydrogen bonding interaction with B increases. As the A....B

Figure 5.2. Qualitative Potential Energy curves for free (———) and hydrogen bonded (-----) A-H group[21].

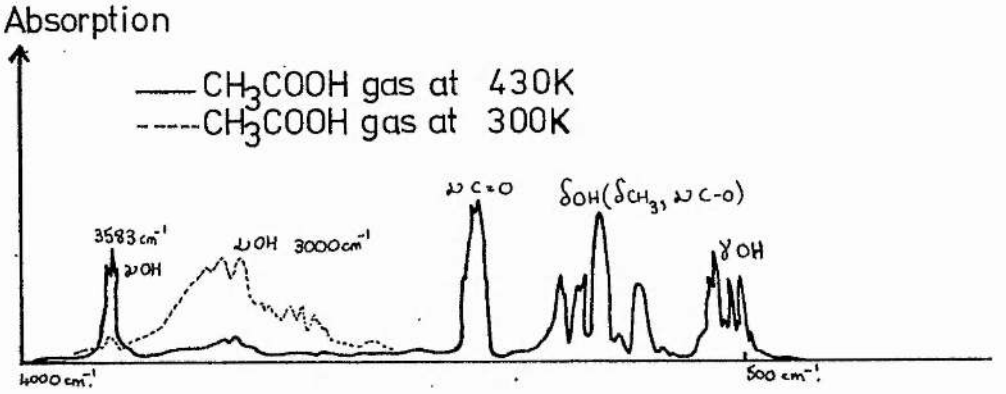


The single minimum anharmonic potential well of A-H is modified to a double minima when hydrogen bonding with B occurs, the position of H being at (I). Transitions to excited vibrational levels, drawn only schematically, are represented by the vertical arrows.

distance decreases, then strength of the hydrogen bonding passes from weak, through intermediate, to strong. (Please see Figure 5.2).

A fairly typical 'free' infra-red absorption frequency for ν OH will be 3583 cm^{-1}. measured in the infra-red spectrum of gaseous acetic acid at 430 K. The absorption band for ν OH is very narrow in this case. As the temperature is lowered, dimerisation begins to occur and O-H...O hydrogen bonding, by the time the temperature has fallen to 300 K, has lowered the ν OH absorption frequency to approximately 3000 cm^{-1}., and the band is now very much broader[21].

Figure 5.3. Infra-red spectra for gaseous CH_3COOH.



An approximate classification of O-H...O hydrogen bond strengths has been given by Novak and is reproduced below, Table 5.4.

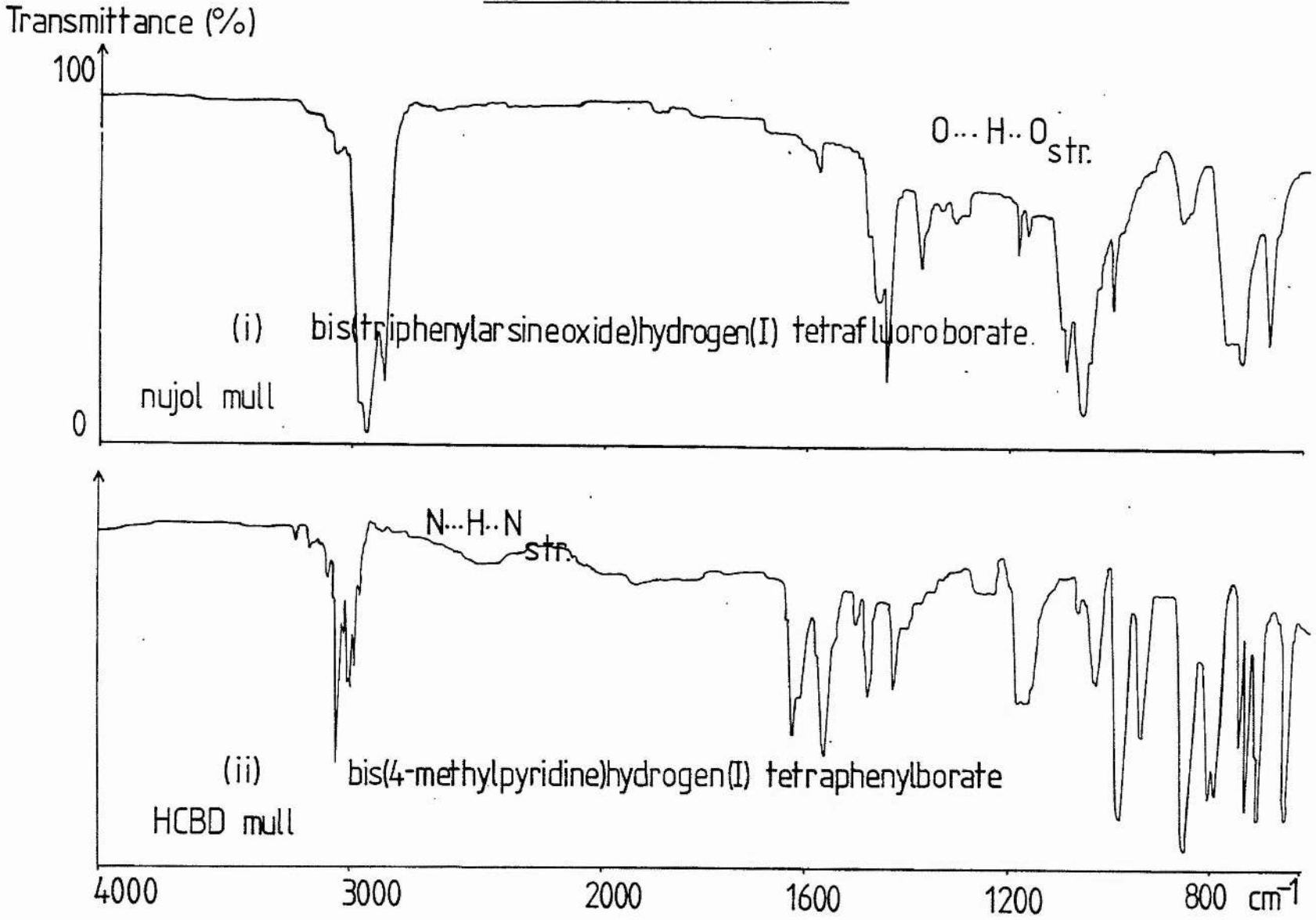
Table 5.4. Approximate classification of O-H...O hydrogen bonds.

Hydrogen bond	ν OH(cm^{-1}) frequency	O....O distance(Å)	Examples
weak	>3200	2.70	H_2O; R-OH ices, hydrates phenols, alcohols
intermediate	3100-2800	2.70-2.60	RCOOH, acids
strong	2700-700	2.60-2.40	$MH(RCOO)_2$, acid salts

According to these criteria, the O...O distance of 2.436(21) Å falls well into the realm of strong hydrogen bonding. The infra-red spectrum of bis(triphenylarsineoxide)hydrogen(I) in a Nujol mull (Figure 5.4(i)) shows that no sharp absorption bands can be directly attributable to νOH, but spanning the region 1600 - 600 cm^{-1}. there is a definite broad shallow absorption.

The bis(triphenylarsineoxide)hydrogen(I) cation does not lie across any special symmetry element. In this work the position of the proton was not located. This is not surprising - positions of hydrogens are usually ill defined in X-ray work, especially when such

Figure 5.4. Infra-red spectra.



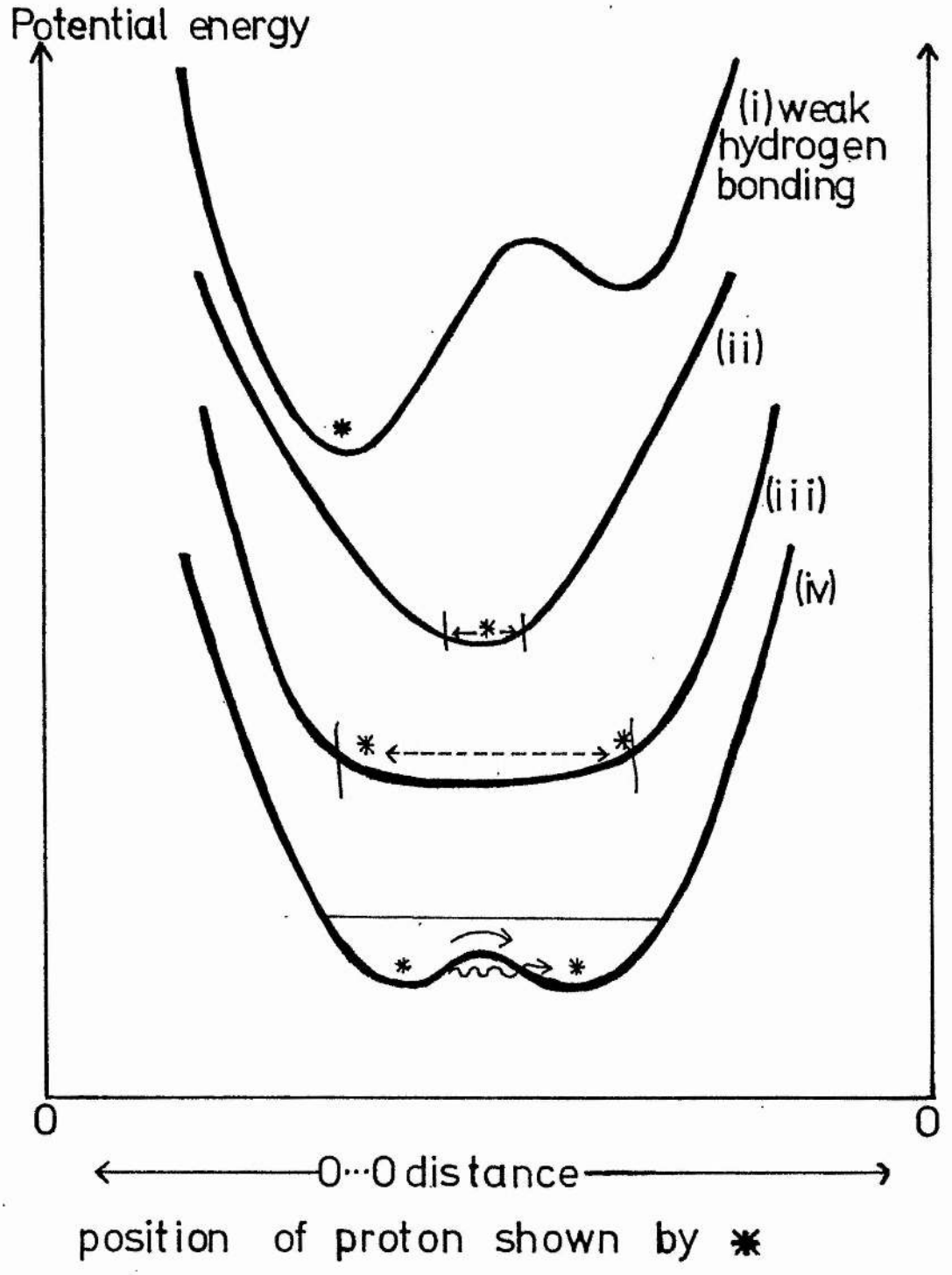
heavy atoms as arsenic are present.

For X-rays, the scattering power of an atom is proportional to $(Z)^2$. Also 'Ripple effects' in the diffraction pattern from these heavy atoms are an aberration caused by the Fourier series not being summed to infinity[22], but can nevertheless mask effectively the very small scattering due to the electron density of any hydrogens, which in any case will be 'spread out' by high thermal motion.

It can reasonably be supposed that in such a short hydrogen bond, gross asymmetry of the type $[A\text{-}O\text{-}H...O\text{-}A]^+$ where A represents the triphenylarsine group is unexpected, that is, that the proton would be associated primarily with one triphenylarsine group, interaction with the other being comparatively weak. This situation generally applies to much longer, weaker hydrogen bonding than is demonstrated in our cation, and a typical potential energy well would be illustrated very schematically by Figure 5.5(i).

The probable situations for strong hydrogen bonding are when the proton is resting at the bottom of a single minimum potential energy well (the widths of this potential energy curve may vary, being either broad, flat bottomed, or rising steeply from the minimum (ii) and (iii) subsequent degree of motion of the proton reflecting this) or, alternatively, the proton may be disordered between two sites (iv). If this happened, it would occupy a site slightly nearer one ligand than the other with a small energy barrier between, and if the two ligands involved in the hydrogen bonding are the same, the potential

Figure 5.5. Potential Energy wells for O...H...O Bonding.



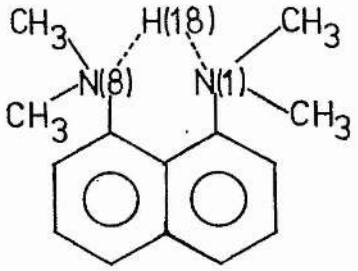
energy curve will be approximately symmetrical. This small energy barrier will offer few constraints to proton motion, which will be disordered between the two energy minima either by passing over the energy barrier or by quantum mechanical tunnelling through it. However, X-ray diffraction work is certainly not able to give any indication of whether the hydrogen bond is an asymmetrical one, with a disordered proton, or symmetrical, the differences being theoretical rather than practically observable[23].

Far fewer very short N...H...N distances have been documented than short O...H...O distances. Some are shown in Table 5.5, which has been taken from Novak's[21] review on hydrogen bonding in solids.

Table 5.5.

Compound	R(N...N)(Å)	νNH frequency(cm^{-1}.)
Hydrazine	3.19	3317
Formamidoxime	3.12	3300
9-methyladenine	3.00	3100
Hydrazinium chloride	2.95	2929
Imidazole	2.86	2800
Purine	2.85	2730
Hydrazinium hydrogen oxalate	2.85	2660
Triazole	2.82	2720

In the 1,8-bis(dimethylammonium)naphthalene cation[24] with tris(hexafluoroacetylacetonato)magnesium or copper (II) as counter ion there is strong N(1)...N(8) hydrogen bonding.



	Cu(II)	Mg
Distance(Å)		
N(1)...N(8)	2.65(2)	2.60(1)
Angle(°)		
N(1)-H(18)-N(8)	148(12)	134(8)

The nitrogen atoms lie on either side, the hydrogen approximately in the plane, of the naphthalene ring. The hydrogen appears closer to N(1) in the copper, to N(8) in the magnesium compound, and the geometry of the hydrogen bond is leading to 6-membered ring formation.

Similar, but less dramatic frequency lowering from the value for the stretching mode in the 'free' N - H bond is found, and also broadening of the absorption band as for O...H...O. The infra-red spectrum for bis(4-methylpyridine)hydrogen(I) tetraphenylborate is illustrated in Figure 5.4(ii). and shows the absorption for N...H...N centred at ca. 2500 cm^{-1}. The N...N distance at 2.610(15) Å is considerably shorter than any quoted above.

Although in this case the bis cation does indeed lie across a centre of inversion, and an electron density peak was located in the difference map at 1/4, 3/4, 1 exactly on this centre of inversion with a small calculated isotropic thermal parameter, this does not give a definite indication that this hydrogen is lying in a single minimum potential well. Speakman[23] has stated that calculated electron densities at special positions are often misleading since these are frequently the sites of anomalously high electron density peaks, or conversely, troughs.

Indeed, the whole question of whether symmetrical hydrogen bonding will ever be realised has been raised[23], theoretical calculations having shown that one will never expect an absolutely symmetrical N...H...N system at N...N distances less than 2.44 Å [25]. The equivalent for the O...H...O system according to Donohue[26] will be an O...O distance of 2.30 Å, although this is an admittedly old estimation, made in 1952.

Detailed neutron diffraction work has shown the hydrogen positions clearly (and to be slightly displaced towards one of the ligands) when X-ray diffraction and symmetry considerations would have indicated centrosymmetry. Displacement of one hydrogen a little way from a crystallographic special position will have such a small effect, that the crystallographic space-group of higher symmetry may be safely adhered to.

Towards the limit, if the energy of the first vibrational level of a double minimum potential energy well is actually higher than the energy barrier, the system will essentially be a flat bottomed single minimum (please see Figure 5.5(iv)).

Such small theoretical differences, although interesting, are rather tangential to the original object of this X-ray work: finding the geometries of these bis cations.

Chapter Five.

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Chapter Six.

The Structures of a Disilazane and a mu-Nitrido-bis (triphenylphosphorus)(I) salt.

I. 1,1,1,3,3,3-Hexaphenyldisilazane.

Preparation.

Triphenylsilylazide was prepared from triphenylsilylchloride and sodium azide following the method of Thayer and West[1]. 1.2 g. of Ph_3SiN_3 and 1.0 g. of Ph_3SiH (commercially available, not further purified) were heated together at 240° C. for 1 1/2 to 2 hours[2]. The yellow oil obtained was taken up in hexane and filtered; after drying the solution with anhydrous Na_2SO_4, the hexane was evaporated off to give a pale yellow oil, which was then chromatographed on alumina with 40/60 petroleum. Elution of the column gave a small ammount of white solid, m.pt. 220° C. which contained no nitrogen (C,H,N analysis).

However, refluxing the alumina from the column with hot petroleum using a Sohxlet extractor, and then evaporating off the petrol, gave a clear residue from which, on standing several days, well-formed, large, clear colourless crystals were obtained, m.pt. 164° C. (literature[2] value for $Ph_3SiNHSiPh$ is 175° C.). Mass spectral

analysis, and C, H, N analysis also strongly supported this compound being $Ph_3SiNHSiPh_3$. Crystals suitable for X-ray work were grown from 40/60 petroleum.

Crystal Data.

$C_{36}H_{31}NSi_2$ is orthorhombic, space-group $P2_12_12_1$ (D_2^4, No. 19). a = 20.072(6), b = 13.317(8), c = 11.249(17) Å, U = 3006.9 Å^3. Mr = 533.82, Dc = 1.179 g.cm^{-3}. for Z = 4. F(000) = 1128.00. Mo-K$_\alpha$ radiation used with λ = 0.71069 Å, μ(Mo-K$_\alpha$) = 1.06 cm^{-1}.

Data Collection and Structure Solution.

Intensity data for 4486 unique reflections having $2^\circ \leq \Theta \leq 30^\circ$ in the octant +h, +k, +l (l = 0 - 14, $0^\circ \leq \mu \leq 26.247^\circ$) were measured using a Stoe STADI-2 diffractometer in the ω-2Θ scan mode, and a scan speed of $0.0167^\circ s^{-1}$. in ω: backgrounds were measured at each end of the peak scan for a period of (scan time/2), and standard reflections after every fifty reflections. Although Lorentz and polarisation corrections were applied, no absorption corrections were.

Space-group $P2_12_12_1$ was uniquely determined by the systematic absences: h00, h ≠ 2n; 0k0, k ≠ 2n; 001, l ≠ 2n.

The structure solution using direct methods, and the following least-squares refinement, were carried out using SHELX 76[3].

465 reflections with normalised structure factors E > 1.4 gave 5163 triple-phase relations which were used to calculate a convergence map. The origin was defined by assigning phases of 90° to reflections 17 3 0 and 11 0 5 and a phase of 0° to 0 8 7. Reflection 7 13 6 fixed the enantiomorph by assigning it a phase angle of 45° or 135°; and reflections 11 8 2 and 18 5 8 were each given phases of 45°, 135°, 225° 315° to yield 32 starting phase permutations. The best E-map from the multi-solution tangent refinement found suitable positions for the two silicon atoms, the nitrogen atom, and nine of the carbon atoms.

Thereafter, blocked full-matrix least-squares refinement with complex neutral atom scattering factors for all atoms[4,5] was carried out, using 2224 unique reflections having $F_o \geq 6\sigma(F_o)$. Anisotropic thermal parameters were applied to Si, N, C, a common isotropic thermal parameter for all phenyl hydrogens (placed in calculated positions) and another isotropic thermal parameter for the unique hydrogen H(1) (whose bonding distance to N was fixed at 1.0 Å), so that in the final cycles of refinement 357 parameters were varied, that is, 120 positional coordinates, 234 anisotropic parameters, one common isotropic parameter, an individual isotropic thermal parameter for H(1) and an overall scale factor. The decrease in Rg was significant at the 99.5 % level for all stages in the refinement[6].

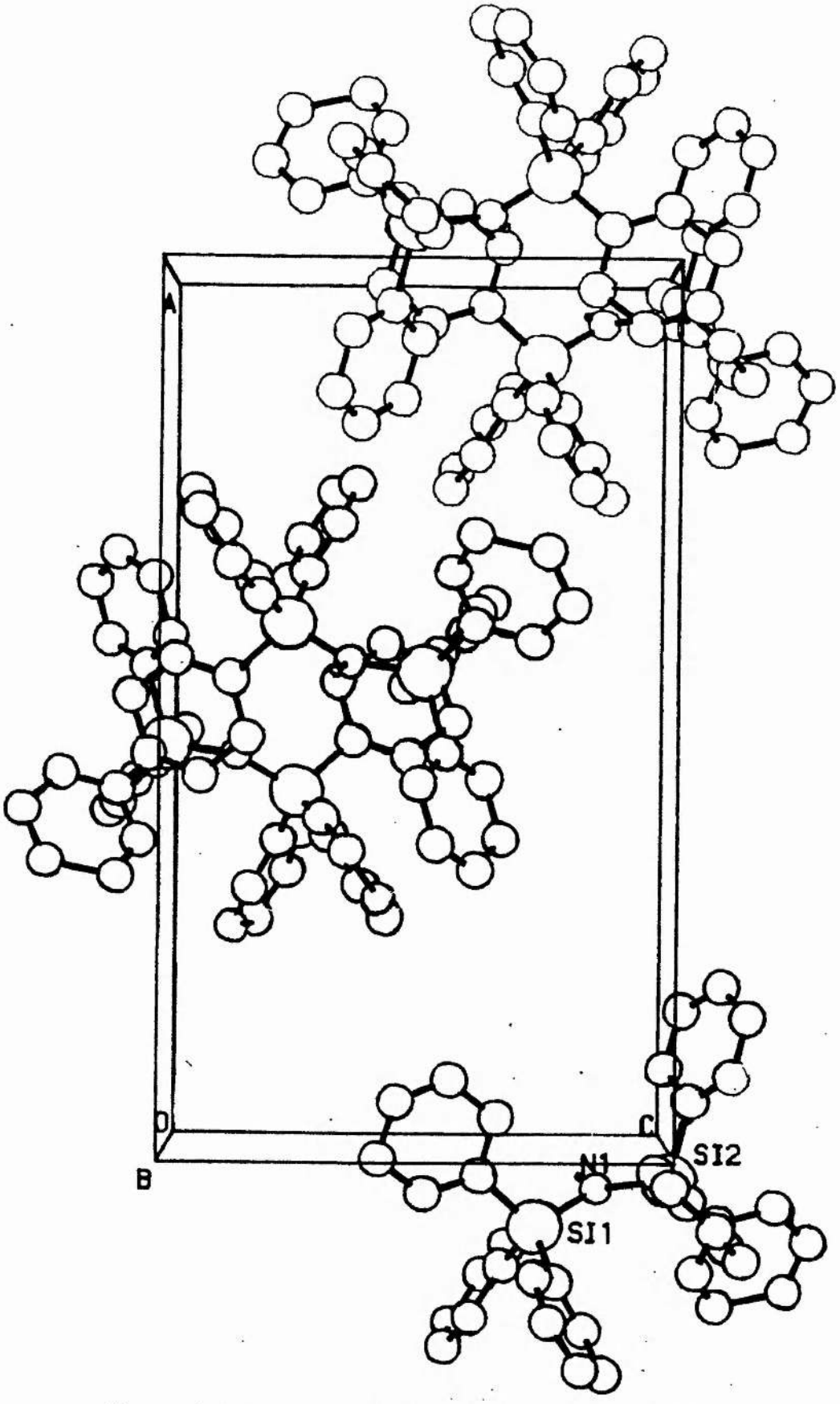

Figure 6.1.2. The contents of the unit cell, viewed down b.

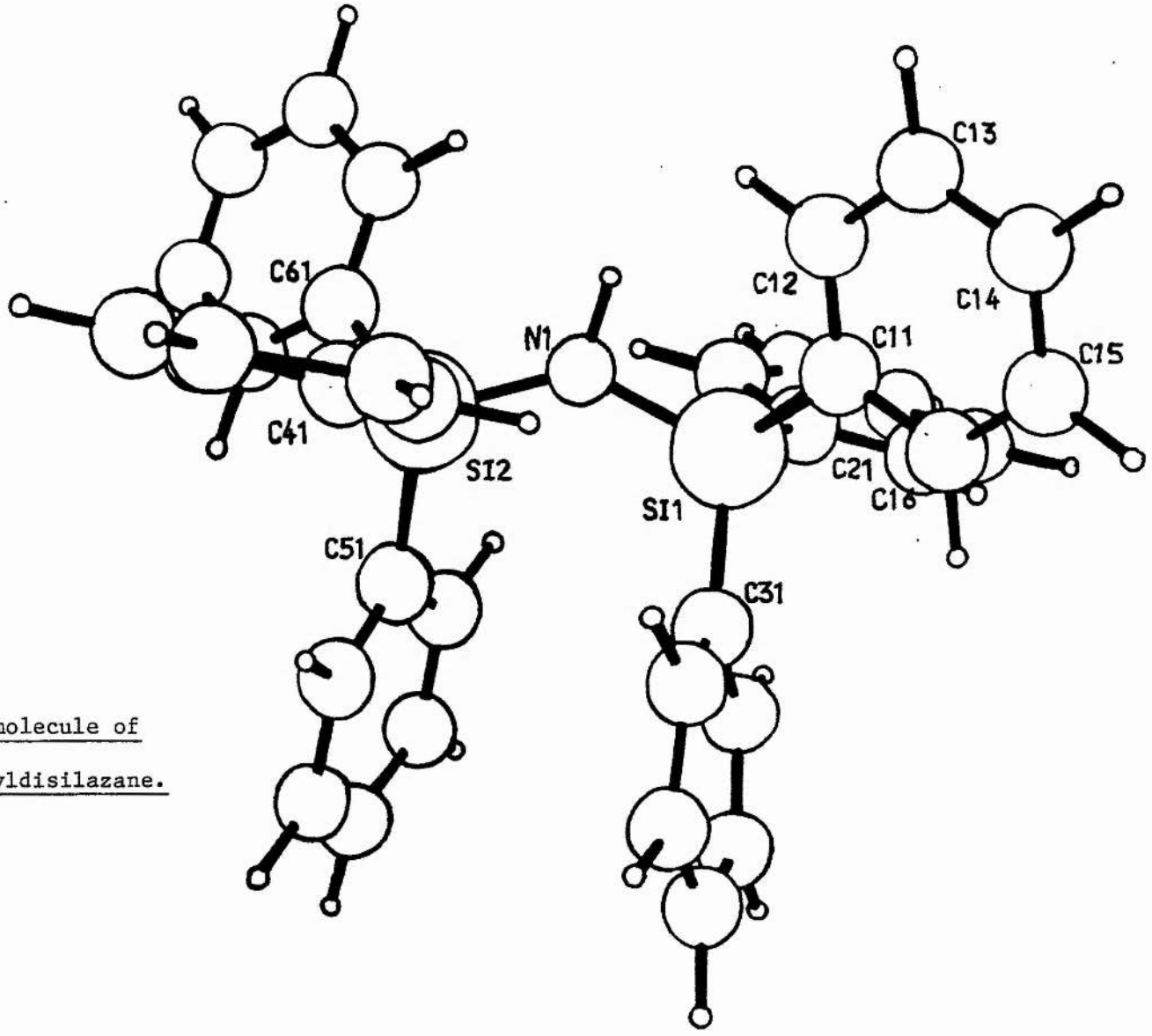

Figure 6.1.1. The molecule of 1,1,1,3,3,3-hexaphenyldisilazane.

Table 6.1.1. Fractional Atomic Coordinates(x 10^4; for H x 10^3).

Atom.	x	y	z
Si(1)	9044(1)	3556(1)	7445(2)
Si(2)	9584(1)	3448(2)	10127(2)
N(1)	9469(4)	3119(5)	8660(7)
C(11)	8553(4)	2500(6)	6776(8)
C(12)	8749(5)	1493(7)	6905(10)
C(13)	8379(5)	722(7)	6406(14)
C(14)	7804(5)	927(8)	5760(10)
C(15)	7590(5)	1904(7)	5631(10)
C(16)	7961(4)	2667(6)	6155(9)
C(21)	9654(4)	4039(6)	6309(9)
C(22)	10321(5)	4180(9)	6617(11)
C(23)	10753(7)	4616(13)	5720(15)
C(24)	10527(9)	4820(11)	4624(18)
C(25)	9858(7)	4660(9)	4257(13)
C(26)	9442(5)	4271(8)	5183(10)
C(31)	8451(4)	4581(6)	7836(8)
C(32)	7891(4)	4393(7)	8506(11)
C(33)	7434(6)	5164(9)	8767(14)
C(34)	7523(5)	6101(8)	8328(11)
C(35)	8073(6)	6301(7)	7694(13)
C(36)	8546(5)	5554(7)	7434(13)
C(41)	9054(4)	2610(5)	11110(8)
C(42)	8465(4)	2233(6)	10679(10)
C(43)	8050(5)	1638(7)	11402(12)
C(44)	8229(5)	1423(7)	12526(12)
C(45)	8826(6)	1786(7)	13007(11)

C(46)	9242(4)	2375(7)	12274(9)
C(51)	9380(4)	4795(6)	10421(9)
C(52)	8990(5)	5091(7)	11186(10)
C(53)	8774(6)	6098(9)	11446(14)
C(54)	9140(6)	6826(8)	10907(12)
C(55)	9616(6)	6551(7)	10047(13)
C(56)	9742(5)	5518(6)	9786(10)
C(61)	10484(4)	3268(6)	10540(8)
C(62)	10842(4)	2492(7)	10043(11)
C(63)	11512(5)	2363(9)	10379(12)
C(64)	11815(4)	2988(8)	11179(10)
C(65)	11458(5)	3761(7)	11638(11)
C(66)	10796(4)	3898(7)	11314(9)

H(1)	-42(9)	243(6)	838(17)
H(12)	-81(1)	133(1)	740(1)
H(13)	-147(1)	-5(1)	652(1)
H(14)	-246(1)	31(1)	533(1)
H(15)	-287(1)	205(1)	515(1)
H(16)	-222(1)	343(1)	606(1)
H(22)	50(1)	398(1)	750(2)
H(23)	129(1)	471(1)	587(2)
H(24)	84(1)	522(1)	399(2)
H(25)	-32(1)	480(1)	337(2)
H(26)	-108(1)	414(1)	498(1)
H(32)	-220(1)	364(1)	884(1)
H(33)	-300(1)	500(1)	932(2)
H(34)	-284(1)	668(1)	849(1)
H(35)	-186(1)	706(1)	735(2)

H(36)	-101(1)	573(1)	691(2)
H(42)	-168(1)	238(1)	977(1)
H(43)	-241(1)	135(1)	1106(1)
H(44)	-209(1)	97(1)	1307(1)
H(45)	-103(1)	162(1)	1391(2)
H(46)	-29(1)	265(1)	1261(1)
H(52)	-142(1)	453(1)	1160(1)
H(53)	-159(1)	633(1)	1210(2)
H(54)	-92(1)	761(1)	1114(1)
H(55)	-12(1)	713(1)	955(1)
H(56)	10(1)	530(1)	912(1)
H(62)	60(1)	199(1)	940(1)
H(63)	180(1)	174(1)	998(1)
H(64)	232(1)	286(1)	1141(1)
H(65)	170(1)	426(1)	1228(1)
H(66)	52(1)	450(1)	1173(1)

Table 6.1.2. Geometry of the $C_3SiNSiC_3$ fragment.

(a) Distances(Å).

Si(1)...Si(2)	3.209(3)		
Si(1) - N(1)	1.718(8)	Si(2) - N(1)	1.722(8)
Si(1) - C(11)	1.874(8)	Si(2) - C(41)	1.896(8)
Si(1) - C(21)	1.883(9)	Si(2) - C(51)	1.870(9)
Si(1) - C(31)	1.864(8)	Si(2) - C(61)	1.881(8)

(b) Angles(°).

Si(1) - N(1) - Si(2)	138.1(4)

N(1) - Si(1) - C(11)	109.1(4)	N(1) - Si(2) - C(41)	109.5(4)
N(1) - Si(1) - C(21)	109.5(4)	N(1) - Si(2) - C(51)	112.6(4)
N(1) - Si(1) - C(31)	112.2(4)	N(1) - Si(2) - C(61)	109.4(4)
C(11) - Si(1) - C(21)	109.0(4)	C(41) - Si(2) - C(51)	109.8(4)
C(21) - Si(1) - C(31)	109.0(4)	C(51) - Si(2) - C(61)	106.8(3)
C(31) - Si(1) - C(11)	108.0(4)	C(61) - Si(2) - C(41)	108.7(4)

II. μ-Nitrido-bis(triphenylphosphorus)(I) thiocyanate.

Preparation.

This compound was prepared by simple exchange of the chloride ion of commercially available μ-nitrido-bis(triphenylphosphorus)(I) chloride, by adding an excess of potassium thiocyanate solution[7]. Recrystallisation was from an ethanol and water mixture to obtain block-shaped crystals, suitable for X-ray diffraction.

Crystal Data.

$C_{37}H_{30}P_2N_2S$ is monoclinic, space-group $P2_1/c$ (C_{2h}^5, No. 14). a = 10.836(3), b = 12.744(5), c = 24.051(4) Å, β = 103.33(2)°, U = 3231.70 Å^3. Mr = 596.66, Dc = 1.226 g.cm^{-3}. for Z = 4. F(000) = 1248.00. Graphite monochromatised Mo-K$_\alpha$ radiation used, λ = 0.71069 Å. μ(Mo-K$_\alpha$) = 1.85 cm^{-1}.

Data Collection and Structure Solution.

Data collection was carried out on a Stoe STADI-2 two-circle diffractometer, using graphite monochromatised Mo-K$_\alpha$ radiation, and an ω-2θ scan mode with a scan speed of 0.0167°s^{-1}. in ω. μ settings took values from 0° to 28.296° for layers k = 0 - 17. Intensities for 4627 reflections were measured, standards being measured every fifty reflections and showing only small random

deviations from their mean intensities. Backgrounds were measured at each end of the peak scan for a time of (scan time/2). Then Lorentz and polarisation corrections were made, but since μ(Mo-Kα) is small, 1.86 cm^{-1}, there were no corrections for absorption.

Systematic absences were: h01, $l \neq 2n$; 0k0, $k \neq 2n$, thus suggesting space-groups $P2_1/c$ or $P2/c$ (C_{2h}^4, No. 13). Structure solution and refinement were successful in $P2_1/c$ (C_{2h}^5, No. 14).

SHELX 76[3] was used to solve and thereafter to refine the structure. The structure was solved in $P2_1/c$ by direct methods using 397 reflections with normalised structure factors $E > 1.4$, giving 5999 triple-phase relations which were then used to calculate a convergence map. The origin was specified by assigning phases of 0^o to reflections -9 1 3, 4 1 14 and 8 2 9. The enantiomorph was fixed by assigning a phase of 45^o or 135^o to reflection -8 4 4; reflections -5 2 17 and -8 2 5 were each given starting phases of 45^o, 135^o 225^o 315^o to calculate 32 starting phase permutations. The best solution from the multi-solution tangent refinement gave an E-synthesis which found suitable positions for the P-N-P fragment and 12 carbon atoms.

After structure solution, refinement of the structure using blocked full-matrix least-squares refinement continued satisfactorily in $P2_1/c$: when positions for all atoms had been found, with P(1), N(3), P(2) given anisotropic thermal parameters and all other atoms isotropic thermal parameters R was reduced to 0.1822 and Rg to 0.2111. At this stage, electron density peaks were beginning to

appear ar feasible positions for phenyl hydrogens and geometries of these rings were acceptable. However, it also appeared that the SCN^- anion was disordered in the crystal since electron density peaks were calculated in the difference map which suggested this ie. as in Figure 6.2.1 and Figure 6.2.2, with the sulphur atom acting as the ´anchor point´ for two disordered molecules; that is, packed into the crystal in two different orientations. By giving the sulphur a site occupation factor (s.o.f.) of one and allowing the s.o.f.´s of the disordered carbon and nitrogen atoms (denoted C1, N1, C2, N2) to refine from 0.5, R and Rg were further reduced to 0.1379 and 0.1435 respectively. It is worthwhile to note that the s.o.f.´s for C1 and C2 refined to 0.60 and 0.54, whereas those for N1 and N2 refined to 0.94 and 0.96, so more electron density than is being accounted for must be present at the positions of N1 and N2. If these s.o.f.´s were used and anisotropic thermal parameters allowed for all atoms of the SCN^- anion, the C and N atoms went non-positive definite and the bond lengths S - C and C - N in the disordered SCN^- anions were now unacceptable. Residual electron density peaks are also seen in the difference map associated with the disordered anion.

This suggests that there is a ´double disorder´, in that whilst the SCN^- anions may take up one of two orientations with respect to the sulphur atom as anchor point, so also may the anions lie ´back to front´ with the sulphur atoms in some cases preferring to occupy approximately the positions of N1 and N2 with their nitrogens lying approximately at the position of S1. However, all attempts to unravel this disorder, thereby obtaining meaningful s.o.f.´s, were

unsuccessful.

For the final cycles of refinement, the phenyl hydrogens were placed in calculated positions and all atoms except S1, P1, P2, N2 (which were allowed anisotropic thermal parameters) given isotropic thermal parameters. 198 parameters were varied comprising 132 positional coordinates, 24 anisotropic thermal parameters, 1 common isotropic thermal parameter for the phenyl hydrogens and one overall scale factor. Complex neutral atom scattering factors were used throughout[4,5].

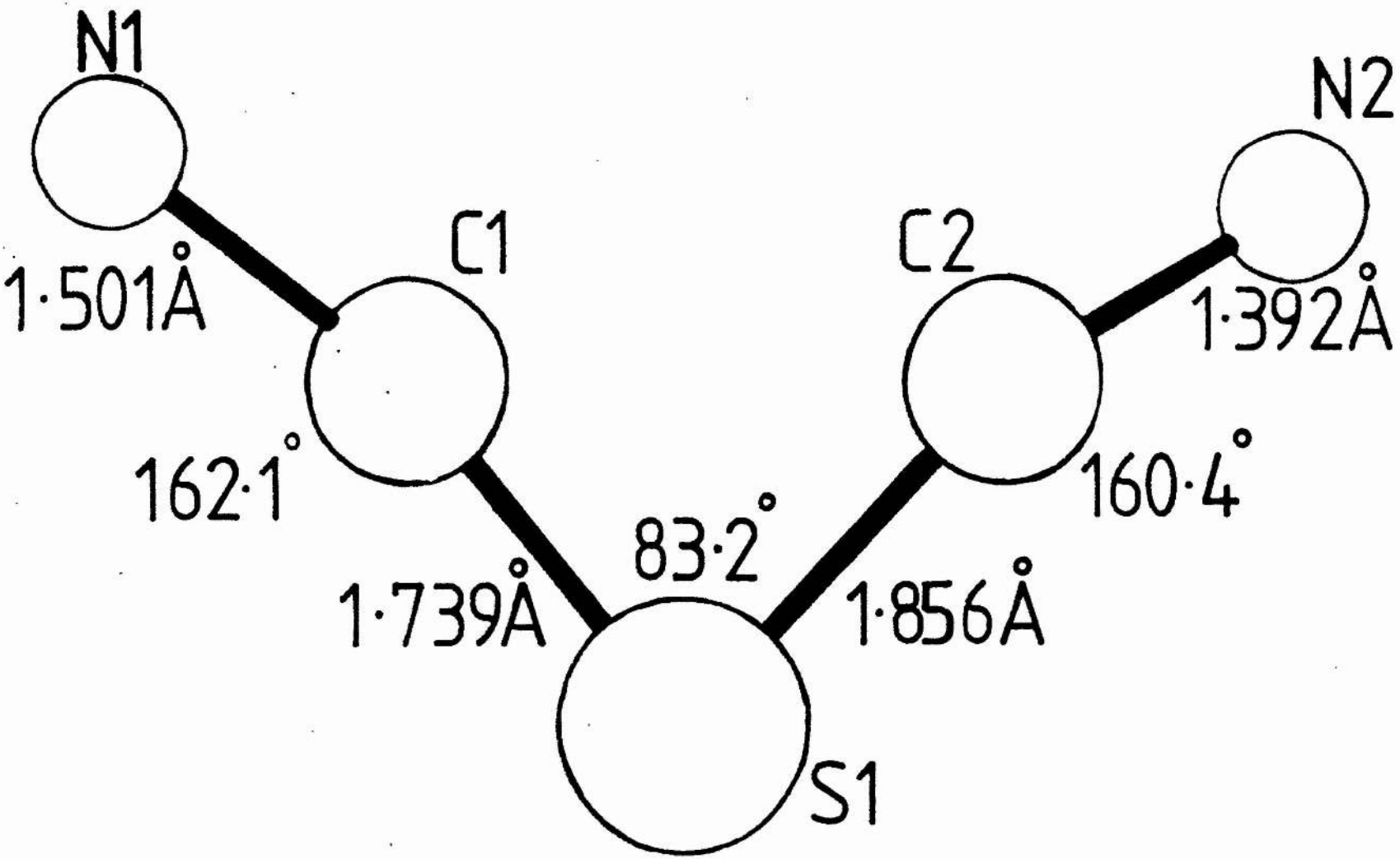

Figure 6.2.1. The single disorder of the SCN^- anion.

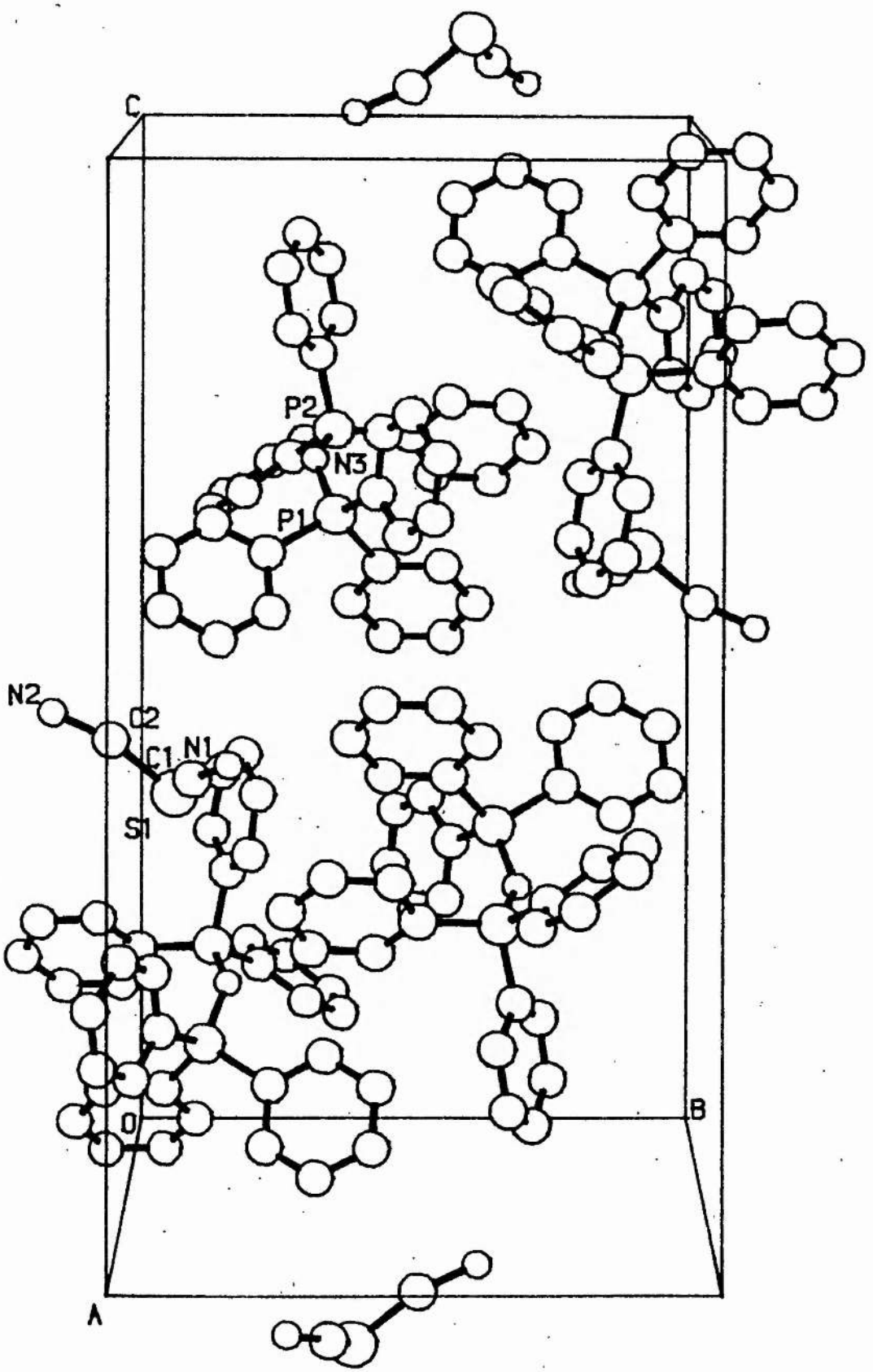

Figure 6.2.3. The unit cell. The view is down a.

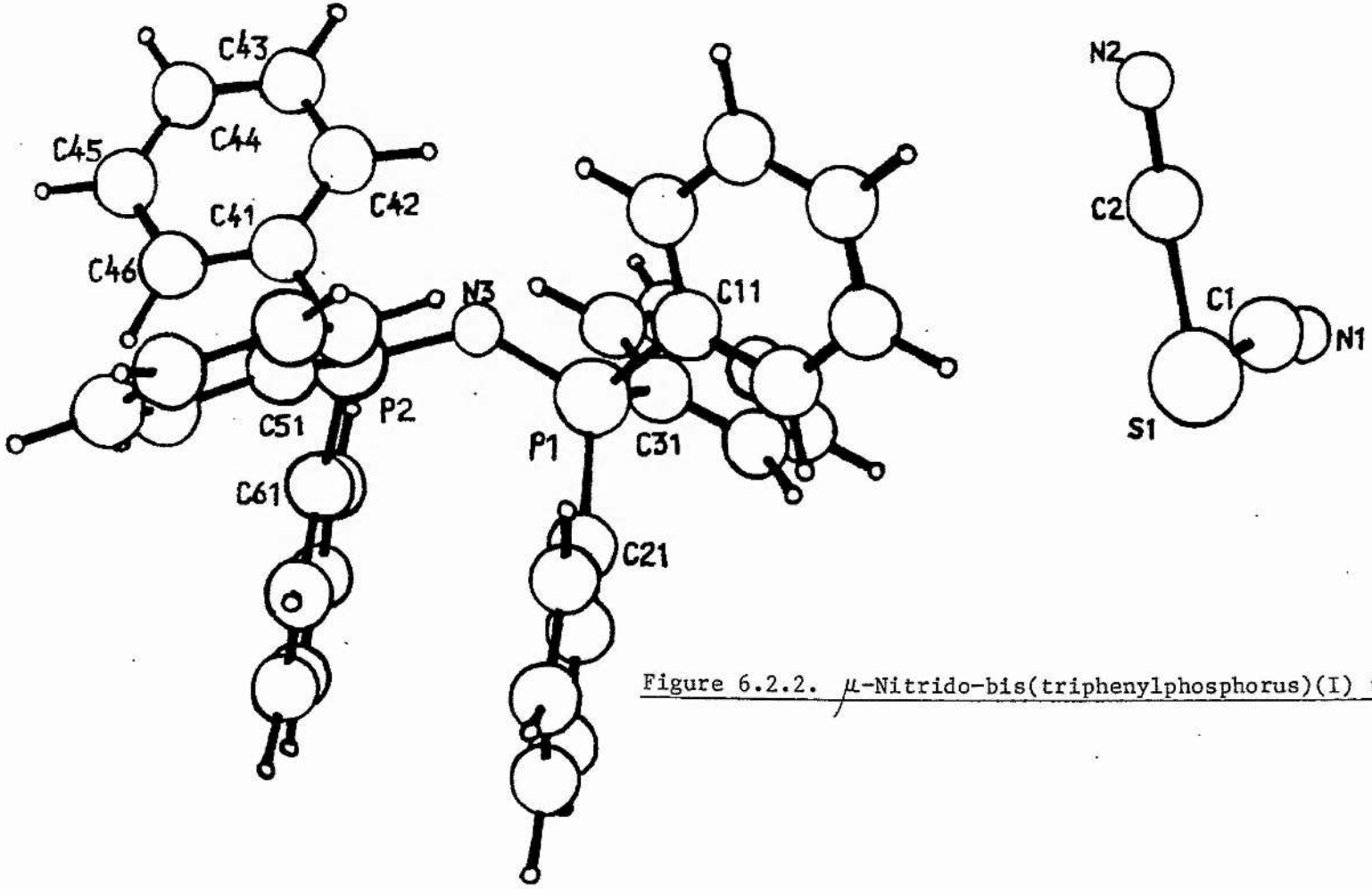

Figure 6.2.2. μ-Nitrido-bis(triphenylphosphorus)(I) thiocyanate.

Table 6.2.1. Fractional Atomic Coordinates(x 10^4; for H x 10^3).

Atom	x	y	z
S(1)	7347(4)	995(4)	4100(2)
C(1)	8892(28)	1371(27)	4415(12)
N(1)	10033(20)	2026(19)	4671(9)
C(2)	7693(27)	-81(26)	4633(12)
N(2)	7690(12)	-1070(11)	4878(5)
P(1)	4389(2)	3629(2)	6394(1)
P(2)	2414(2)	3565(3)	7052(1)
N(3)	3670(7)	3254(8)	6866(3)
C(11)	4919(8)	2467(8)	6079(4)
C(12)	4958(9)	1523(9)	6358(4)
C(13)	5407(10)	631(10)	6128(5)
C(14)	5788(11)	705(11)	5620(5)
C(15)	5745(10)	1669(10)	5344(5)
C(16)	5314(10)	2548(10)	5565(5)
C(21)	3458(10)	4410(9)	5824(4)
C(22)	2556(11)	3874(11)	5398(5)
C(23)	1755(13)	4481(12)	4987(6)
C(24)	1841(13)	5556(13)	4989(6)
C(25)	2704(13)	6064(13)	5395(6)
C(26)	3527(11)	5487(11)	5829(5)
C(31)	5785(9)	4351(9)	6713(4)
C(32)	6564(11)	4795(10)	6382(5)
C(33)	7672(12)	5320(11)	6639(6)
C(34)	8028(13)	5423(12)	7220(6)

C(35)	7279(13)	4948(12)	7556(6)
C(36)	6136(11)	4463(10)	7306(5)
C(41)	2675(9)	3287(8)	7807(4)
C(42)	3795(10)	2863(9)	8102(4)
C(43)	4012(12)	2708(11)	8702(5)
C(44)	3063(12)	2999(11)	8978(6)
C(45)	1937(12)	3420(11)	8675(5)
C(46)	1723(11)	3612(10)	8084(5)
C(51)	1093(9)	2776(8)	6683(4)
C(52)	-154(12)	3013(11)	6736(5)
C(53)	-1138(13)	2357(12)	6460(5)
C(54)	-910(13)	1515(12)	6135(6)
C(55)	313(12)	1313(12)	6091(6)
C(56)	1317(10)	1928(9)	6356(4)
C(61)	1987(9)	4920(9)	6968(4)
C(62)	1083(10)	5317(10)	6503(5)
C(63)	864(11)	6389(11)	6441(5)
C(64)	1558(12)	7089(12)	6826(5)
C(65)	2487(14)	6717(13)	7287(6)
C(66)	2699(13)	5637(12)	7350(6)
H(12)	464(1)	147(1)	675(1)
H(13)	545(1)	-11(1)	635(1)
H(14)	613(1)	2(1)	544(1)
H(15)	605(1)	172(1)	495(1)
H(16)	528(1)	329(1)	535(1)
H(22)	249(1)	303(1)	540(1)
H(23)	105(1)	410(1)	466(1)
H(24)	121(1)	600(1)	466(1)

H(25)	277(1)	691(1)	539(1)
H(26)	421(1)	588(1)	616(1)
H(32)	629(1)	473(1)	592(1)
H(33)	826(1)	564(1)	638(1)
H(34)	888(1)	585(1)	742(1)
H(35)	759(1)	498(1)	801(1)
H(36)	553(1)	416(1)	757(1)
H(42)	451(1)	265(1)	788(1)
H(43)	489(1)	238(1)	894(1)
H(44)	320(1)	287(1)	943(1)
H(45)	121(1)	362(1)	890(1)
H(46)	86(1)	398(1)	785(5)
H(52)	-33(1)	368(1)	698(1)
H(53)	-209(1)	252(1)	650(1)
H(54)	-169(1)	102(1)	592(1)
H(55)	49(1)	64(1)	585(1)
H(56)	226(1)	177(1)	630(1)
H(62)	56(1)	478(1)	619(1)
H(63)	13(1)	667(1)	609(1)
H(64)	139(1)	792(1)	677(1)
H(65)	303(1)	726(1)	759(1)
H(66)	342(1)	535(1)	770(1)

Table 6.2.2. Geometry of C_3PNPC_3.

(a) Distances(Å).

P(1) ... P(2)	2.942(4)				
P(1) - N(3)	1.592(9)	P(2)	-	N(3)	1.579(9)

P(1) - C(11)	1.817(10)	P(2) - C(41)	1.807(10)
P(1) - C(21)	1.802(10)	P(2) - C(51)	1.806(9)
P(1) - C(31)	1.786(10)	P(2) - C(61)	1.787(12)

(b) Angles(°).

P(1) - N(3) - P(2)	136.3(6)		
N(3) - P(1) - C(11)	107.8(5)	N(3) - P(2) - C(41)	106.9(4)
N(3) - P(1) - C(21)	115.4(5)	N(3) - P(2) - C(51)	111.0(5)
N(3) - P(1) - C(31)	110.7(5)	N(3) - P(2) - C(61)	115.4(5)
C(11)- P(1) - C(21)	108.0(5)	C(41)- P(2) - C(51)	107.9(5)
C(11)- P(1) - C(31)	106.3(5)	C(41)- P(2) - C(61)	106.3(5)
C(21)- P(1) - C(31)	108.2(5)	C(51)- P(2) - C(61)	109.1(5)

Table 6.2.3. Geometry of the disordered Anion.

(a) Distances(Å).

S(1) - C(1)	1.739(28)	S(1) - C(2)	1.856(32)
C(1) - N(1)	1.501(37)	C(2) - N(2)	1.392(36)

(b) Angles(°).

S(1) - C(1) - N(1)	162.1(25)
S(1) - C(2) - N(2)	160.4(21)
C(1) - S(1) - C(2)	83.2(14)

Chapter Six.

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Chapter Seven.

Bending of Molecular Pseudohalides.

I. Introduction.

The term ´pseudohalide´ was first introduced by Birkenbach and Kellerman to describe polyatomic groups which resemble halides in their chemical properties, such as -NNN (azide), -CNO (fulminate), -NCO (cyanate) and -NCS (thiocyanate)[1].

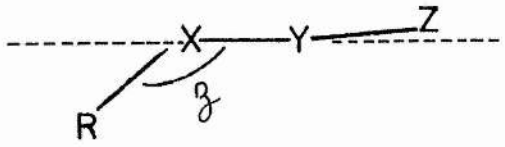
MNDO[2] calculations have been carried out on various pseudohalides: only a little background theory and results of such calculations are reported here, mention of most of the theoretical basis being reserved until the following chapter, where results will be discussed with the crystal structures reported in Chapter Six, along with other crystal structures investigated in recent years, many by Glidewell and Liles, as data for commenting upon the ´Second Order Jahn-Teller Effect´.

II. Optimum Geometries.

Pseudohalide molecules RXYZ were investigated where -XYZ is the pseudohalide group, and R is H, F, Cl or a C- or Si- containing group with various permutations of H, F and Cl attached.

By carrying out SCF calculations using the MNDO programme system[2], the potential energies of these pseudohalides were repeatedly lowered by altering the bonding distances and interatomic angles until a potential energy minimum was reached. Thus the optimum geometry for the pseudohalide in the gas phase was found.

The pseudohalides will have the structure



and it is in angle R-X-Y, θ, that interest lies. One fact is immediately obvious from Table 7.1 - some pseudohalides are linear and some are bent at X. It is found that the XYZ fragment is roughly linear in pseudohalides linear at X, and in those bent, a trans configuration is generally adopted. Two typical examples are drawn in Figure 7.1.

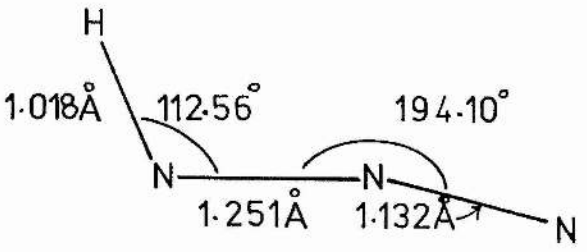
Table 7.1(a). Optimised Geometries for Pseudohalides, RXYZ. (°)

	XYZ =			
R =	-NNN	-NCO	-CNO	-NCS
H-	112.6	120.4	179.9	132.8
F-	113.5	119.0	180.0	122.0
Cl-	*molecule dissociates	116.6	180.0	121.2
CH_3-	119.7	131.1	179.9	153.1
CH_2F-	121.2	134.0	179.9	152.1
CF_2H	120.2	132.8	179.8	152.9
CF_3-	119.3	130.5	179.9	147.4
CH_2Cl	118.8	130.9	177.1	154.5
$CHCl_2$	120.1	132.2	178.5	155.4
CCl_3-	121.6	132.9	179.9	155.9
CF_2Cl	119.2	132.5	177.7	151.9
CCl_2F-	119.7	131.8	178.9	152.0
CHFCl-	120.5	132.5	177.7	153.2

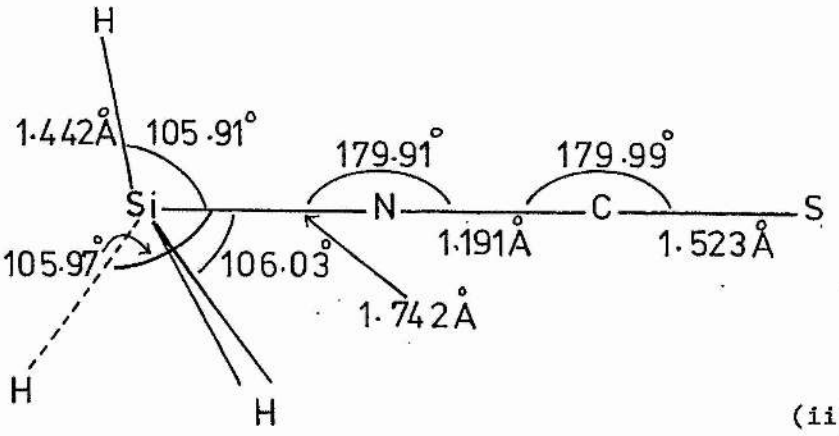
Table 7.1(b).

	-XYZ =		
R =	-NNN	-NCO	-NCS
H_3Si-	124.0	179.9	179.9
H_2FSi-	126.5	172.7	178.8
F_2HSi-	124.5	174.9	178.7
F_3Si-	120.0	179.7	180.0
H_2ClSi-	128.6	170.5	176.6
Cl_2HSi-	127.1	174.7	177.8
Cl_3Si-	123.9	179.7	180.0
F_2ClSi-	124.2	175.8	176.7
Cl_2FSi-	123.4	178.6	178.5
HFClSi-	126.9	173.2	176.3

Figure 7.1. Bent and linear pseudohalide molecules - optimised geometries.



(i) HNNN



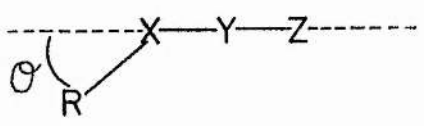
(ii) H_3SiNCS

Although the N_3^- ion is symmetrical and linear, HN_3 and CH_3N_3 have been investigated[3], and have angles H-N-N and C-N-N of 110° and 135°. As expected, calculated geometries of covalent azides are very bent with γ approximately 110-130° for both C- and Si- bonding to the pseudohalide moiety.

For -NCO and -NCS, the molecules are bent with C attached, but approximately linear with γ in the range 170-180° for R groups containing Si. Also as expected, for -CNO the molecules are linear: the Si-bonded analogues have not been investigated using MNDO, since presumably, they too will be linear.

III. Potential Energy Profiles.

For a given pseudohalide, how does the energy of the molecule vary with θ, the angle from linear geometry at X?



θ was fixed at certain angles 0°, 10°, 20°.....90°, and a series of MNDO calculations were carried out for each molecule. Whilst θ was fixed, other bond lengths and interbond angles were optimised during successive SCF calculations, so minimising the potential energy of the molecule at each θ value. Thus enthalpies of formation calculated by

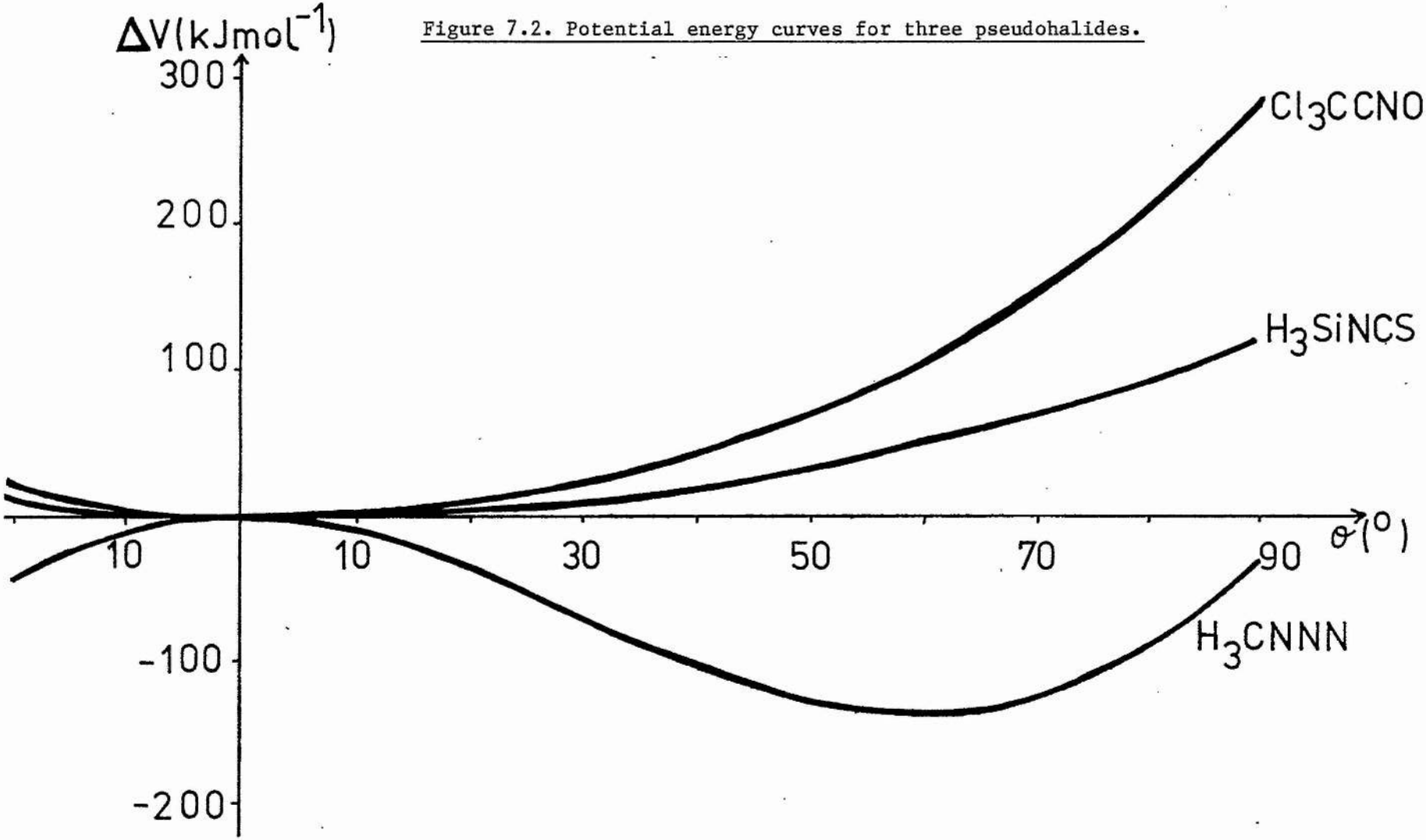


Figure 7.2. Potential energy curves for three pseudohalides.

MNDO will measure the potential energy of the molecule, and curves can be drawn of calculated ΔH_f° against fixed θ values, to show energy variation with bending of the molecular geometry. All potential energy curves are necessarily symmetrical about the linear geometry.

The next stage is to find the energy differences for angles of θ, with the potential energy for the linear molecule, $P.E._{\theta} - P.E._{linear}$, ie. ΔV.

The potential energy curves can be described by a polynomial having the form:

$$\Delta V = C + a\theta^2 + b\theta^4 + c\theta^6 + d\theta^8 \ldots\ldots$$

where θ has now been converted from degrees to radians. C is a constant and is the point at which the calculated curve intercepts the y axis, which plots ΔV values. Since ideally, this is at $\Delta V = 0$, the smaller the value of C, the more accurate is the fitting of the polynomial expansion to the actual curve. Because the potential energy curves are symmetrical, only even powers of θ are necessary for the polynomial. Using a polynomial expansion programme written by Mr. A.F. Cuthbertson[4], ΔV and θ^2 values are fitted to the above polynomial. It is the first coefficient of the polynomial, that corresponding to θ^2, which is needed for calculation of the force constant for bending of the linear geometry.

Consider a simple harmonic oscillator in a potential well: the potential energy of the oscillator, U, is

$$U = 1/2\ kx^2$$

where x is the displacement of a particle from the minimum of the well, and k is the restoring force constant.

Differentiating

$$\frac{dU}{dx} = kx$$

$$\text{and } \frac{d^2U}{dx^2} = k$$

The second derivative of the potential energy function therefore gives the force constant.

Since, in exactly the same manner, the potential energy curve, or 'well', for bending of the linear pseudohalide can be described thus:

$$\Delta V = C + a\theta^2 + b\theta^4 + c\theta^6 + d\theta^8 \ldots\ldots$$

Differentiating gives

$$\frac{\partial \Delta V}{\partial \theta} = 2a\theta + 4b\theta^3 + 6c\theta^5 + 8d\theta^7 \ldots\ldots$$

$$\text{and } \frac{\partial^2 \Delta V}{\partial \theta^2} = 2a + 12b\theta^2 + 30c\theta^4 + 56d\theta^6 \ldots\ldots$$

Therefore, since the interest lies in bending of the linear geometry, ie. at θ = 0 radians

$$\left(\frac{\partial^2 \Delta V}{\partial \theta^2}\right)_{\theta = 0} = 2a$$

because all other terms contained powers of θ and are now 0. So, twice the calculated coefficient 'a' from the calculated polynomial will give the curvature of the potential energy well for linear geometry (units are $kJmol^{-1}rad^{-2}$). The conversion factor

$$\frac{2a}{l_1 l_2 N_A}$$

(where l_1 and l_2 are the bond lengths R - X and X - Y for the linear geometry and N_A is Avogadro's Number) is employed to convert this into the more conventional force constant, with unit Nm^{-1}.[5] Values are tabulated in Table 7.2.

Table 7.2(a). Force Constants for bending of linear geometries (Nm^{-1}).

	XYZ =			
R =	-NNN	-NCO	-CNO	-NCS
H-	-101.028	-28.079	13.208	-8.132
F-	* molecule dissociates	-79.175		-40.920
Cl-	*molecule dissociates	-32.575	0.206	-15.294
CH_3-	-71.336	-16.155	8.738	-2.273
CH_2F-	-92.485	-25.992	9.725	-7.679
CF_2H-	-102.679	-24.064	11.106	-4.381
CF_3-	-62.912	-13.257	12.576	-2.839
CH_2Cl-	-92.779	-27.919	4.872	-10.124
CCl_2H-	-89.965	-22.740	10.100	-4.815
Cl_3C-	-64.167	-13.065	12.962	-1.812
F_2ClC-	-61.484	-15.116	19.331	-6.117
Cl_2FC-	-61.371	-14.706	6.509	-2.000
HFClC-	-91.670	-22.000	5.393	-9.051

Table 7.2(b).

	-XYZ =		
R =	-NNN	-NCO	-NCS
H_3Si-	-12.566	1.946	4.259×10^{-3}
H_2FSi-	-19.833	-1.654	4.929×10^{-3}
F_2HSi-	-15.433	-2.036	4.895×10^{-3}
F_3Si-	-12.124	3.333	4.150×10^{-3}
H_2ClSi-	-24.423	-3.856	6.908×10^{-3}
Cl_2HSi-	-24.105	-3.582	6.857×10^{-3}
Cl_3Si-	-10.371	4.028	3.936×10^{-3}
F_2ClSi-	-15.845	8.077	8.226×10^{-3}
Cl_2FSi-	-21.935	1.930	4.961×10^{-3}
HFClSi-	-21.924	-1.036	2.743×10^{-3}

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Chapter Eight.

The Second Order Jahn-Teller Effect and Molecular Geometries.

I. Some 'Unexpected' Molecular Geometries.

In 1955, Hedberg[1] presented the result of a gas phase electron diffraction study of trisilylamine, which indicated that the molecule had an essentially coplanar heavy atom Si_3N skeleton and a shorter than expected Si - N bonding distance. Beagley et al.[2] repeated this work in 1970, and the results were identical: Si - N, 1.736(2) Å, angle Si-N-Si, 119.1(1)°, Si....Si, 2.998(3) Å. This coplanar geometry has been used to rationalise the chemical observation that trisilylamine is a weak electron donor (compared to pyramidal trimethylamine which is a very strong Lewis base) in terms of $p_\pi - d_\pi$ delocalisation of the lone pair of electrons from the $2p_z$ orbital of N into low lying empty d orbitals of the silicons. Electron diffraction work on $(H_3Si)_3P$[3] and $(H_3Si)_3As$[4] shows that the heavy atom skeletons are pyramidal. If one advocates back bonding into empty Si 3d orbitals to explain the geometry of the trisilylamine, then presumably the lone pair in the $3p_z$ and $4p_z$, orbitals of P and As respectively, are too high in energy and are too diffuse for efficient overlap to occur in a $p_\pi - d_\pi$ delocalised system, and in these cases the lone pairs of electrons retain their

stereochemical activity.

Trigermylphosphine is also pyramidal, from infra-red and Raman spectra: two skeletal bending modes observable in the Raman give strong evidence of a non-planar geometry[5].

Another silicon containing compound with an 'unexpected' geometry, which has aroused much interest, is disiloxane, $(H_3Si)_2O$. Gaseous disiloxane was studied in the gas phase by electron diffraction[6] and the Si-O-Si angle is large, 144.1(8)°, very much larger than would be expected from a simple VSEPR[7](Valence Shell Electron Pair Repulsion) consideration and from a comparison of other analogues. Gaseous disilyl sulphide, $(H_3Si)_2S$, also studied by electron diffraction[8], has the expected very bent geometry with an Si-S-Si angle of 97.4(7)°.

Low temperature single crystal X-ray work has been carried out for disiloxane (108K) and hexamethyldisiloxane (148K)[9]. Si-O-Si angles were found to be 142.2(3) and 148.8(2)°, and Si - O distances 1.631(5) and 1.626(2) Å respectively. The Si....Si distances are 3.086(2) and 3.132(1) Å. Also in accord with this X-ray work, is an electron diffraction study of hexamethyldisiloxane[10], for which an Si - O distance 1.631(3) Å and Si-O-Si angle 148(3)° were calculated.

Whenever electron diffraction studies are carried out for molecules in the gas phase, there is always the possibility that the molecule is not being studied in its ground state, the structure instead representing the average over all thermally populated vibrational levels. On the other hand, if a molecule is studied in the solid state and has an easily bent skeleton, then intermolecular forces may have become important, thereby altering the molecular geometry from that of the 'isolated' ground state. A different, more efficient, packing arrangement may lower the potential energy of the crystalline substance. For molecules which may have very easily deformed skeletons, what is really needed is a very low temperature, electron diffraction study, if this becomes possible, so that neither intermolecular forces nor thermally excited states have any effect on the molecular structure.

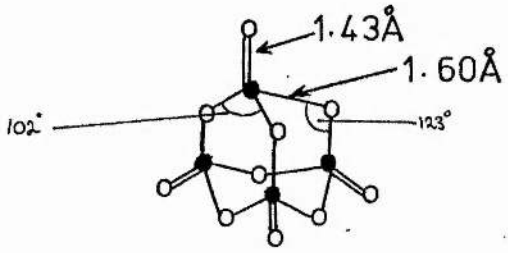
Other halogenated silicon ethers, including $(F_3Si)_2O$[11] and $(Cl_3Si)_2O$[12] have been studied by electron diffraction, their large Si-O-Si angles being determined as 155.7 and 146° respectively.

II. 'Very Short' bonds involving Silicon.

When the first electron diffraction studies of $(H_3Si)_3N$[1] and $(H_3Si)_2O$[6] were carried out, why should the results have caused such a great deal of discussion? Cruickshank in 1961[13] discussed the role of 3d orbitals in $p_\pi - d_\pi$ bonding between (Si, P, S or Cl) and (O or N) to explain many unexpected geometries. In particular, backbonding,

and hence an increase in the strength of the bond, was used to rationalise some 'very short' bonds, and the very large angles which tend to be found in many silicon containing compounds. But were the bonds really so 'very short'? Pauling[14] suggested lengths using values of covalent radii, and Schomaker and Stevenson[15] then corrected these by taking into account values of electronegativities for the atoms involved (a concept which is nebulous to say the least, and virtually impossible to place on a numerical scale). All distances for Si - O bonds, although much shorter than those suggested by Pauling or Schomaker and Stevenson are nevertheless remarkably constant at approximately 1.63 Å, showing very little variation with the groups attached. If $p_\pi - d_\pi$ bonding were important then a much greater variation would be expected - consider P_4O_{10} which has formally been represented as (I). The difference in bond lengths is about 0.2 Å[16].

(I)



Dismiss the idea of such Si - O bonds being unusually short, considering instead 1.63 Å to be a perfectly normal single bond length, then $p_\pi - d_\pi$ bonding need no longer be used to 'explain' "unusual" geometries in silicon compounds. Another approach, originally suggested by Pearson[17,18], makes use of the 'Second Order Jahn-Teller Effect' to rationalise molecular geometries.

III. The Second Order Jahn-Teller Effect.

This approach takes into account the symmetry of a species, and examines its stability with respect to distortion along a normal coordinate.

> The Second Order Jahn-Teller Theorem says that any system whose ground state symmetry is $[P_o]$ which has a low lying excited electronic state of symmetry $[P_1]$, will distort along a non-totally symmetric normal vibration, whose symmetry is contained in $[P_o] \times [P_1]$.

Let the potential energy of a molecule in its ground state be E_o. Then perturb this geometry a little along a normal vibrational coordinate Q, so that the molecular symmetry is altered. The energy for the molecule, still in its ground electronic state, can be written as[17]

$$E = E_o + \frac{1}{2} V_{oo} Q^2 + \sum_k \frac{V_{ok}^2 Q^2}{E_o - E_k} \qquad (1)$$

E_o is the energy of the unperturbed molecule, and the E_k's those of various excited electronic states. In practice, unless the first several excited states lie energetically very close together, only the first is important in the summation.

$$V_{oo} = \int \psi_o \left(\frac{\partial^2 V}{\partial Q^2}\right) \psi_o \, d\tau$$

and

$$V_{ok} = \int \psi_o \left(\frac{\partial V}{\partial Q}\right) \psi_k \, d\tau \quad .$$

in which ψ_o is the ground state wave function, ψ_k an excited state wave function and V the nuclear-nuclear and nuclear-electronic potential energy.

Another way of writing equation (1) is[19]

$$E = E_o + 1/2 \; f_{oo}Q^2 + 1/2 \; f_{ok}Q^2 \qquad (2)$$

Now, the sum of f_{oo} and f_{ok} is the experimental force constant for perturbation of the molecule. The first term is the classical force constant, giving the restoring force back towards the original electron and nuclear distribution. It is always positive. The second term, conversely, is always negative and gives a reduction in the restoring force - it has been called the 'relaxability' of the molecule along a coordinate, Q[19].

If the value of $1/2 \; f_{ok}Q^2$ is sufficiently <u>negative</u>, the value of E will be less than of E_o. Thus the potential energy of the perturbed ground state species will be <u>lower</u> than of the unperturbed species. The next question is: which conditions are necessary for the making of

$1/2\ f_{ok}Q^2$ into a large negative term, thereby making the perturbed species much more stable? Looking at equation (1), if the value of $E_o - E_k$ is small, then this will help the condition to be satisfied. If the energy gap between the HOMO (Highest Occupied Molecular Orbital) and the LUMO (Lowest Unoccupied Molecular Orbital) is small, perturbation of the molecule will be spontaneous, ie. when there is a low lying excited state.

The nature of the HOMO and LUMO must also be considered. If two orbitals are very close together in energy, but the direct product of their symmetries does not contain the symmetry of a normal vibration, then distortion will not be possible. In addition, the condition that the vibration must be non-totally symmetric is included in the theorem: otherwise, the distortion would cause no outward change.

This may be thought as a 'blending together' of the HOMO and LUMO when distortion occurs.

There are three possibilities for the value of $(f_{oo} + f_{ok})$, the force constant f_Q for perturbation along a normal coordinate, Q:

(i) It may be large and positive - then the molecule will remain rigidly unperturbed. Any distortions along Q will cause a large increase in the molecule's potential energy.

(ii) It may be negative - the molecule will then distort until it has a nuclear and electronic arrangement for which the potential energy is minimised.

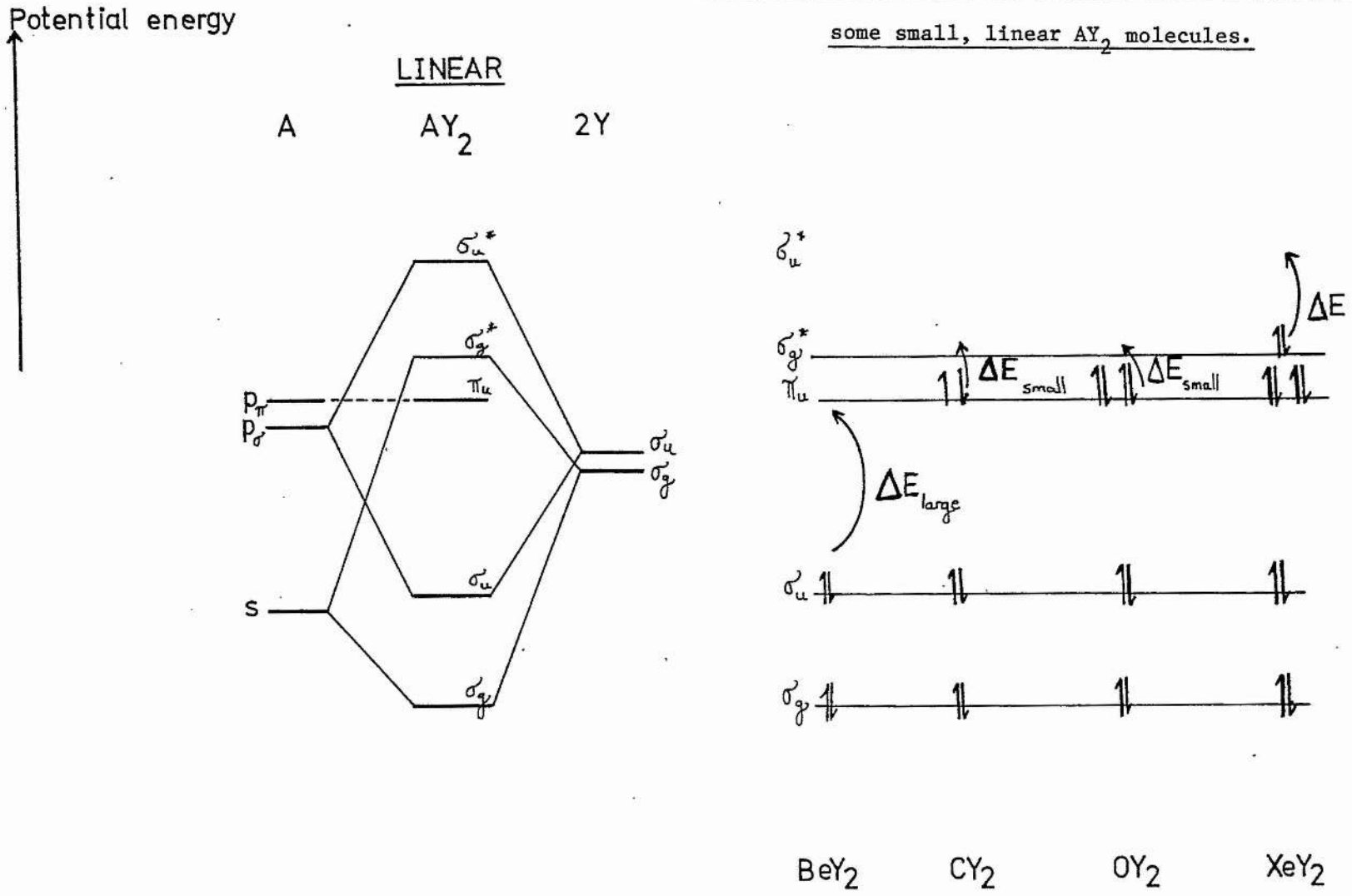
(iii) f_Q may be approximately zero - in this case a very broad, flat-bottomed potential energy curve might be expected, large perturbations of the molecular geometry causing only very small differences in the potential energy of the molecule.

IV. Some Examples.

The previous section has dealt (in a very descriptive, qualitative manner) with the basic ideas of the Second Order Jahn-Teller Effect. Some examples of its application are now given.

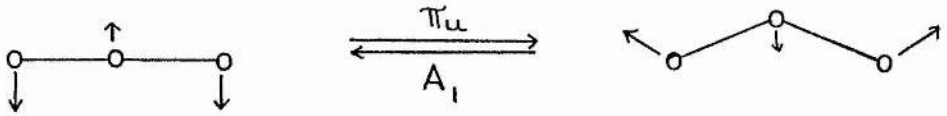
Taking the very simple example of linear AY_2 species: a molecular orbital correlation diagram for such simple species would be shown schematically in Figure 8.1[20]. In the case of BeY_2, where Y is a monodentate ligand or atom, there will be 4 electrons involved in σ bonding, occupying the σ_g and σ_u molecular orbitals. The energy gap between the σ_u and π_u orbitals is large, so the linear configuration will be stable to bending. CY_2 and OY_2 both have the non-bonding π_u orbital as their HOMO. The energy gap between the π_u and σ_g^* energy levels is now small, so the possibility of Jahn-Teller distortion away

Figure 8.1. Molecular orbital correlation diagrams for some small, linear AY_2 molecules.



from the linear form will now arise.

Taking the product of $[\pi_u]$ x $[\sigma_g^*]$, there is a normal vibrational coordinate, of symmetry $[\pi_u]$ contained in this product. This is a bending mode,



and so the likely distortion of the molecule is bending from the linear geometry. The A_1 vibrational mode will take the bent AY_2 species back to linearity. According to this theory, a carbene CY_2 should be bent, at least in the singlet state, and molecules such as H_2O, Cl_2O, F_2O, all having 8 electrons involved in bonding, are all bent, with angles at oxygen of 104.5, 110.8 and 103.2°, respectively[20].

In the above, only the σ bonding in a molecule has been considered. If p_π (or d_π) orbitals have the correct symmetry for interaction and are at suitable energy levels, they also must be taken into account.

Ligand electronegativity, we must also expect to affect molecular geometries, since this will alter the relative energy gaps between the HOMO and LUMO, and hence the value of the complete term $1/2\ f_{ok}Q^2$.

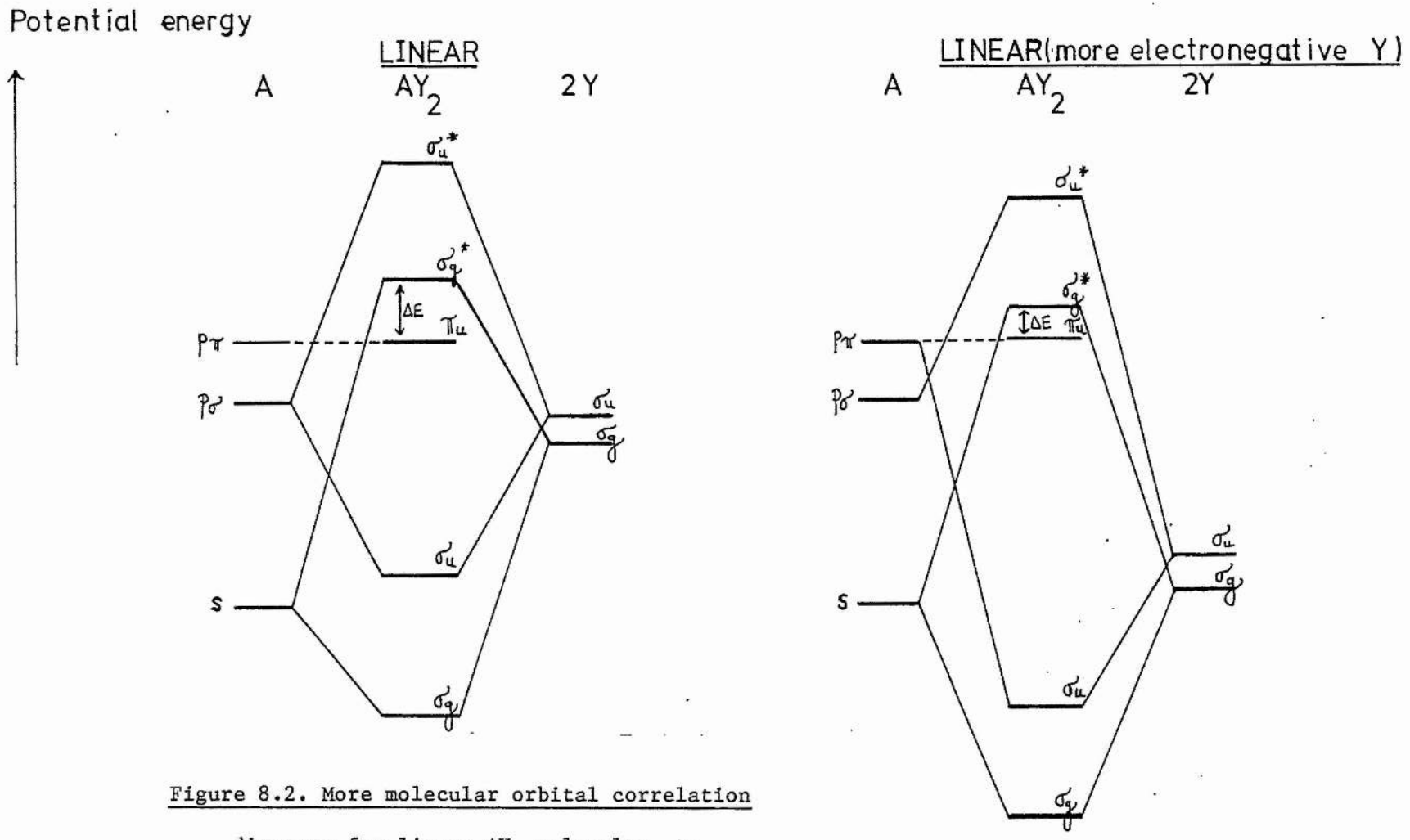


Figure 8.2. More molecular orbital correlation diagrams for linear AY_2 molecules, to illustrate effect of ligand electronegativity.

Consider again linear AY_2, in Figure 8.2. Figure 8.2 is very schematic and exaggerated, but increasing the electronegativity of the Y ligands will lower the potential energies of the interacting orbitals on Y: because the energy of, say, the non-bonding π_u remains the same (call this the HOMO) then the $\pi_u - \sigma_g^*$ energy gap may be lowered enough to now allow distortion to occur.

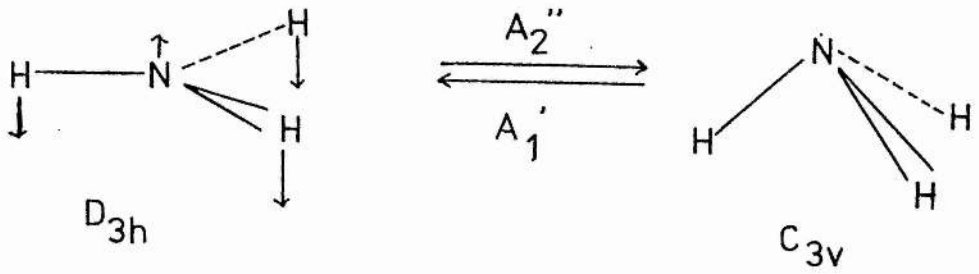
It will be seen that the SOJT effect provides a rationalisation of VSEPR theory (which is concerned with localised bonding and non-bonding electron pairs around a central atom), and Walsh's Rules (a molecular orbital approach), wherein the <u>total</u> number of valence electrons is used to predict the molecular geometry.

V. Distortions of MX_3 species.

Returning to the very first topic of this chapter, planar $(H_3Si)_3N$, its 'unusual' geometry may now be rationalised. The simplest analogue, NH_3, would have a molecular orbital sequence[21]

$$(a_1')^2(e')^4(a_2'')^2(2a_1')^0(2e')^0$$

in the planar D_{3h} form. The excitation from $(a_2'') \longrightarrow (2a')$ is easy, and contained in $[a_2''] \times [a']$ is the vibrational mode $\supset A_2''$. This is the umbrella bending mode converting NH_3 to the pyramidal point group C_{3v}.



A 6-21G ab initio calculation[22], with geometry optimisation, for NH_3 shows the energy of the molecular orbitals to be (in atomic units)

$(a_1)^2$.... $(a_1)^2$	$(e)^4$	$(a_1)^2$	$(a_1)^0$	$(e)^0$
-15.534 -1.126	-0.619	-0.389	+0.274	+0.377
N(1s)		HOMO	LUMO	

The $(a_1) \longrightarrow (a_1)$ energy gap of 0.633 a.u. is large, so distortion of pyramidal to planar ammonia will not occur.

Even if this energy gap were small, $[a_1] \times [a_1]$ only gives the possibility of the totally symmetric A_1 vibration which causes no distortion of the nuclei.

But, for $(H_3Si)_3N$, the energy of the orbital from the H_3Si- moiety involved in the σ' bonding will be very different to that of the H(1s) orbital. In this way, the energy gap between HOMO and LUMO for D_{3h} trisilylamine may be large enough to preclude distortion to the pyramidal form, and so also may the pyramidal form be unstable to the A_1' vibration which will convert the molecule back to planarity.

VI. Easily Deformed AY_2 species.

Disiloxane, and other silicon ethers, were the second compounds whose seemingly 'unusual' geometries were commented upon. An angle at oxygen of ca. 140-150° was considered to be exceptionally large, compared to the approximately tetrahedral angles predicted by VSEPR theory.

Whilst VSEPR successfully predicts geometries for compounds involving first row elements, it fails to predict correctly the geometries for compounds involving first and second row elements. For example, disiloxane. Now, the geometry seems to be governed by the 'close contact' distance. In $R_3SiOSiR_3$ compounds, the Si....Si distance is remarkably constant at about 3.1 Å, indicating that the silicon atoms can go no closer together (ie. the angle at oxygen decrease) without electronic repulsions substantially raising the potential energy of the molecule. By considering atoms as hard, rigid bodies, whose size determines their distance apart, such an approach has been used to calculate covalent radii for several p-block elements[23].

Analogous sulphur compounds had very similar Si....Si distances, but now with angles at sulphur of approximately 100°, as expected[8].

So, the large angles in bent disiloxanes could be explained by a hard atom model. But this does not explain observations of linear Si-O-Si fragments.

Linearity has been observed in several silicates. Cruickshank[24], following early (1930) work by Zachariasen, found Si-O-Si in thortveitite, $Sc_2Si_2O_7$, to be linear. Similar work on rare earth pyrosilicates[25] found the following Si-O-Si angles: $Eb_2Si_2O_7$, 180°; $Yb_2Si_2O_7$, 180°; $Gd_2Si_2O_7$, 158.6°; $Nd_2Si_2O_7$, 132.6°. Another rare earth silicate, $Er_4PbSi_5O_{17}$, which contains both the $Si_2O_7^{6-}$ and $Si_3O_{10}^{8-}$ anions, again has a linear pyrosilicate anion[26].

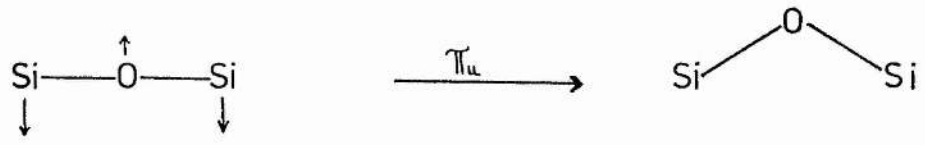
In molecular species, linearity of Si-O-Si was first suggested in hexa-(γ -trifluoropropyl)disiloxane[27] from symmetry requirements. The final R factor was 26 % with only 568 intensity data. Extensive disorder of the trifluoropropyl groups occurs, and possible disordering of oxygen and hence bending of Si-O-Si cannot be discounted. One feels that a linear Si-O-Si skeleton cannot be firmly accepted from this work.

Glidewell and Liles[28] found that in crystals, molecules of $Ph_3SiOSiPh_3$ are centrosymmetric, and so Si-O-Si is required to be strictly linear, the first example of a linear silicon ether with organic substituents. The benzyl analogue, likewise, is linear[29].

Returning to the Second Order Jahn-Teller Effect: a plausible elctronic configuration for an Si-O-Si bridge is[30]

$$(1\sigma_g^+)^2\ (1\sigma_u^+)^2\ (1\pi_u)^4\ (2\sigma_g^+)^0\ (2\sigma_u^+)^0$$

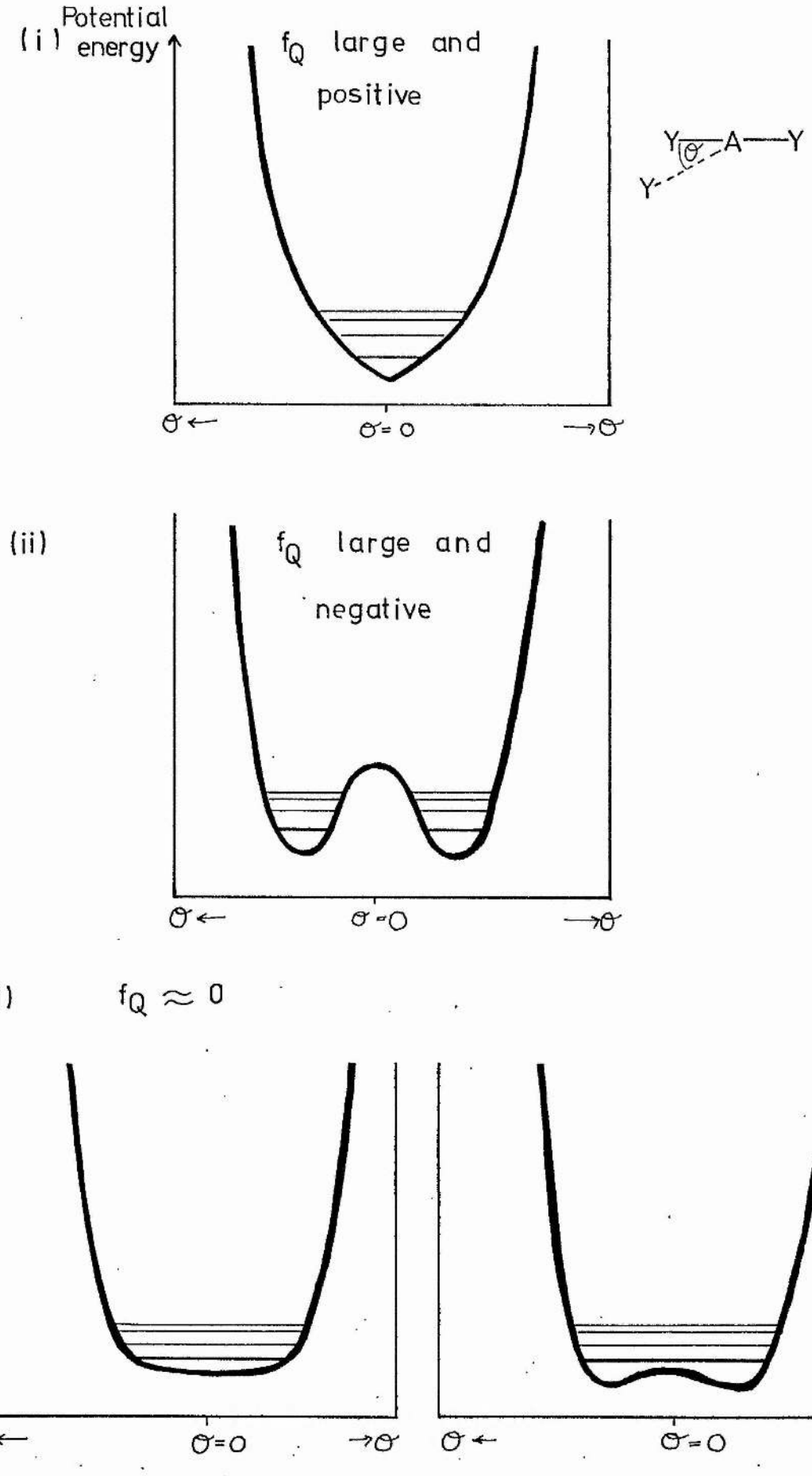
therefore any distortion of the skeleton will be contained in $[\pi_u] \times [\sigma_g^+] = \pi_u$ which is the bending vibration.



Condition (i) in Section (III) (and Figure 8.3) is for a positive force constant for π_u bend. The molecule remains linear. For (ii), a negative force constant, the potential energy of the molecule will be lower after bending. Condition (iii), force constant ≈ 0, will arise when there is a broad, flat potential well.

Potential energy wells for in-plane bending of an AY_2 molecule will be symmetrical about the linear geometry, and are illustrated in Figure 8.3. In the limit, as an energy potential of type (ii) becomes shallower, the first vibrational level may be such that the potential well is virtually flat.

Figure 8.3. Potential energy curves for bending of linear AY_2.



The diversity of angles found in Si-O-Si skeletons suggests that the force constant for the π_u distortion is ≈ 0 (iii). Electron diffraction work has indicated bent Si-O-Si (although for $O(SiF_3)_2$ the possibility of a linear ground state could not be ignored[28].

With dramatic changes in geometry involving only minimal changes in potential energy of the anion or molecule for bending or straightening of the bridge, intermolecular forces will be very important in determining the solid state geometry.

Isoelectronic with an Si-O-Si bridge are P-C-P and $P-N-P^+$ bridges. The crystal structure of Ph_3PCPPh_3 shows there are 2 independent molecules in the unit cell, having P-C-P angles of 130.1^o and 143.8^o. By postulating a very easily deformed 'soft' P-C-P skeleton, intermolecular forces may account for the angles differing by 13.7^o[31].

The X-ray structures of μ-nitrido-bis(triphenylphosphorus)(I) thiocyanate, which was reported in Chapter Six, was undertaken to find how the P-N-P skeleton behaved when a fairly small anion was present as the counter ion. μ-Nitrido-bis(triphenylphosphorus)(I), so called 'PPN' salts are widely used to stabilise many inherently unstable metal cluster compounds and polynuclear carbonyls because of their bulkiness (please see Chapter Five, section V). Many times the structure of the 'PPN' cation has been reported inadvertently, when the main topic of interest was the anion. The range of P-N-P angles

is worthy of comment. The thiocyanate which we examined was bent, P-N-P angle 136.3(6)°, being at the lower end of the range of angles which go to 180° in eg. $[(Ph_3P)_2N]^+[V(CO)_6]^-$[32]. A selection of geometries is given in Table 8.1.

Kirtley et al. have shown the presence of one linear and two bent (139.1°) PPN cations in the same unit cell of $[(Ph_3P)_2N]_3\{Na[Mo_3(CO)_6(NO)_3(\mu_2\text{-}OCH_3)_3(\mu_3\text{-}O)]_2\}$[33].

It seems reasonable to surmise that the bridging skeletons in all of these compounds are readily deformed and that lattice forces and efficient packing in the crystal determine the solid state geometry.

Table 8.1. Some Geometries of PPN salts.

Compound	PNP(°)	d(P-N)(Å)	Ref.
$PPN^+[Fe(CO)_4CN]^-$	134.6(31)	'typical'	34
PPN^+SCN^-	136.3(6)	1.592(9), 1.579(9)	Chapter Six
PPN^+TeCN^-	143.2(3)	1.563(5), 1.578(5)	35
$PPN^+[HOs_5(CO)_{15}]^-$	144.5(10)	1.55(2) 1.59(2)	36
$PPN^+[Cr_2(CO)_{10}(\ -H)]^-$	154.6(3)	1.553(2)	37
$PPN^+[Ni_2(CO)_6H]^-$	175.9(8)	1.549	38
$PPN^+[V(CO)_6]^-$	linear	1.539(2)	32
$3PPN^+\{Na[Mo_3(CO)_6(NO)_3$	Two bent:	1.59(2)	33

	139.1(12)	1.56(2)
$(\mu_2\text{-}OCH_3)_3(\mu_3\text{-}O)]_2\}^{3-}$	One linear	1.53(1)

Further evidence for a very anharmonic, large amplitude bending mode for disiloxane and d_6-disiloxane has been given by infra-red and Raman spectra. This work calculates a low barrier to linearity of only 112 cm^{-1}., and of 95 cm^{-1}. for the d_6-molecule, which are 'in accord with the concept of a quasi-linear molecule'[39].

Similarly, the spectra of $(Cl_3Si)_2O$ suggest that it is bent (C_{2v} selection rules) as a gas, but in the crystalline state, that it straightens (D_{3h} selection rules)[39]. Whilst infra-red and Raman work erroneously suggested $(H_3Si)_3P$[3] and $(H_3Si)_3As$[4] to be planar, it seems very possible that $(Cl_3Si)_2O$, also, has an easily deformed Si-O-Si skeleton, and that packing forces are important.

VII. p_π - p_π Delocalisation?

Ernst et al. have shown that only oxygen d orbitals must be included in an ab initio calculation, not silicon d orbitals to obtain a double minimum potential energy curve. But the height of the 'bump' is very small, less than 1 $kJmol^{-1}$. which agrees well with the 112 cm^{-1}. just mentioned[40].

Glidewell shows, via a 6-31G* ab initio calculation for $(H_3Si)_2O$, that electron density is delocalised from what is essentially the O $2p_x$ lone pair[41]. The coefficients for the silicon $2p_x$ and hydrogen 1s orbitals are considerable, whilst those for silicon 3d orbitals are negligible, thus implying that $p_\pi - p_\pi$ interaction is more important than $p_\pi - d_\pi$ interaction.

Whilst a number of reviews have been written on the subject of $p_\pi - d_\pi$ interaction in silicon-containing compounds[42-45], SOJT distortions, coupled with electron delocalisation via p orbitals only, adequately rationalises some 'unusual' geometries. The energy gained by delocalisation of the lone pair in, for example, trisilylamine, will be unlikely per se to compensate for a much less stable geometry and to cause the molecule to be planar.

Rather, SOJT distortion will change the molecular framework, now making delocalisation of electron density a possible effect. Slight decrease in bond length of Si - N in trisilylamine will be a reflection of the increasing 's' character of the σ bonding orbitals, now ostensibly sp^2 rather than sp^3. P - N distances of 1.59(2), 1.56(2) Å in the bent PPN^+ cation, compared to 1.53(1) Å in the linear[33], also illustrate this trend.

VIII. MNDO work for PPN^+ analogues.

Because the $(Ph_3P)_2N^+$ cation is simply too large for molecular energy calculations to be carried out, MNDO methods were applied to small analogues, viz. $(H_3P)_2N^+$ and $(Me_3P)_2N^+$[46]. Such calculations show that, in contrast to the small value of f_Q for the π_u vibrational mode in PPN^+ cations, both $(H_3P)_2N^+$ and $(Me_3P)_2N^+$ are calculated by MNDO to have substantial barriers to linearity of 85.7 and 75.8 $kJmol^{-1}$.: hence large, negative force constants are implied for the π_u vibration of the linear cations. A value of -87.9 Nm^{-1}. was calculated for $(H_3P)_2N^+$.

IX. ESCA work on PPN^+ salts.

It has been suggested that ESCA studies (photoelectron spectroscopy) of the binding energies of the N(1s) and P(2p) electrons in PPN^+ salts may be a convenient way of determining whether the cation is linear or bent without undertaking a full X-ray analysis. ESCA studies were carried out, to find whether formalism (I) or (II) is most likely[47].

$[(C_6H_5)_3P{=}\overset{+}{N}{=}P(C_6H_5)_3]$ (I)

$[(C_6H_5)_3\overset{+}{P}{-}\overset{-}{N}{-}\overset{+}{P}(C_6H_5)_3]$ (II)

(I) is more likely to be associated with linearity, (II) with bending. The authors concluded[47] that (II) was most appropriate. But the N(1s) and P(2p) binding energies are very insensitive to cation geometry (Table 8.2), with little energy differences for known bent and linear species. (I) would involve electron donation back towards the phosphorus atoms, most favoured when angle P-N-P is 180°. Bonding intermediate between limits (I) and (II) is most likely.

Table 8.2. P(2p) and N(1s) Binding Energies for PPN^+X^- salts.

Anion	P(2p)(eV)	N(1s)(eV)
F^-	132.8(3)	397.6(2)
Cl^-	133.0(2)	397.6(3)
Br^-	133.2(2)	397.4(3)
I^-	132.9(3)	397.5(2)
OCN^-	133.0(2)	397.2(1)
		400.5(2)*
SCN^-	133.3(3)	397.3(3)
		400.2(3)*
NO_3^-	133.2	397.2(2)
N_3^-	133.2	397.2
$V(CO)6^-$	133.3(2)	397.5(2)

* Value for anion nitrogen

Distribution of electron density in P-N-P fragments also seems to be independent of P-N-P angle.

X-ray powder diffraction patterns were recorded for $(Ph_3P)_2N^+X^-$ (X^- = Cl^-, Br^-, I^-, NO_2^-, NO_3^-, NCO^-, NCS^- and N_3^-) using a Philips powder diffractometer: $(Ph_3P)_2N^+F^-$ was extremely hygroscopic, and so no powder diffraction could be recorded for this[46]. Comparison of these powder diffraction patterns showed none of the salts were isomorphous, and none were isomorphous with linear $PPN^+V(CO)_6^-$, which is additional support for P(2p) and N(1s) binding energies being geometry-independent.

X. Hexaphenyldisilazane and similar compounds.

Work on many $(R_2Si)_2X$ compounds has now been carried out, both by electron diffraction for small gaseous molecules, and by single crystal X-ray diffraction. The geometries of some are listed below, Table 8.3. (Bz = $C_6H_5CH_2$-)

Table 8.3. Geometries of some $R_3SiXSiR_3$ molecules (X = O, NH, CH_2, S, Se).

Compound	Si - Si(Å)	Si....Si(Å)	Si-X-Si(°)	Ref.
$H_3SiNHSiH_3$	1.725(3)	3.097(6)	127.7(1)	48
$Me_3SiNHSiMe_3$	1.735(12)	3.085(49)	125.5(18)	49
$Ph_3SiNHSiPh_3$	1.718(6)	3.209(3)	138.1(4)	50 and Ch. 6
$H_3SiOSiH_3$(108K)	1.631(6)	3.086(2)	142.2(3)	9
$Me_3SiOSiMe_3$(148K)	1.626(5)	3.132(1)	148.8(2)	9
$Ph_3SiOSiPh_3$	1.616(1)	3.232(1)	180.0(0)	28
$Bz_3SiOSiBz_3$	1.613(4)	3.226(4)	180.0(0)	29
$H_3SiCH_2SiH_3$	1.873(2)	3.149(3)	114.4(2)	51
" (MNDO)	1.836	-	116.4	52
$Me_3SiCH_2SiMe_3$(MNDO)	1.858	-	125.5	52
$Ph_3SiCH_2SiPh_3$	1.87	3.336	126.1	53
$H_3SiSSiH_3$	2.136(2)	3.210(19)	97.4(7)	8
$H_3SiSeSiH_3$	2.273(4)	3.395(14)	96.6(5)	54

When X is a first row element, the Si....Si distance is very constant at about 3.1 Å. True, Si....Si distances are longer, and Si - X distances shorter for R = phenyl or benzyl, but the Si-N-Si angle of 138.1(4)° for hexaphenyl- is the largest so far recorded for a disilazane[50]. Similarly, Si-O-Si angles for these substituents are

a maximum and these compounds are linear[28,29]. Work is still in progress on the crystal structure of $Ph_3SiCH_2SiPh_3$[53], but this angle subtended at carbon is much larger than tetrahedral, and larger than that calculated from an electron diffraction study of $H_3SiCH_2SiH_3$[51] or via MNDO calculations[55] with optimisation of geometry for $Me_3SiCH_2SiMe_3$[52]. The Si-C-Si angle of 114.4° in $H_3SiCH_2SiH_3$ is 2° larger than the C-C-C angle in $H_3CCH_2CH_3$[51].

The central C-C-C angle in $Me_3CCH_2CMe_3$ is also opened to 125-128°[56]. An MNDO calculation, with optimisation of geometry, is in total accord with this, predicting the C-C-C angle to be 127.0[52]. Dimethylether and diethylether have angles C-O-C of 111.5 and 108° respectively[57]. For dipropylether, an electron diffraction study shows the C-O-C angle to be rather larger, 116.1(36)°[58].

Considering again, a double minimum potential energy curve for bending of a linear molecule (Figure 8.3). Going towards linearity from the bent optimum geometry will be energetically unfavourable. Similarly, a more bent molecule will have a higher potential energy because the energy rises sharply when nuclear-nuclear and electronic repulsions become important. In bent molecules, the 3.1 Å is probably the 'close-contact' distance of the two silicons, closer than which these two atoms repel one another strongly. Bulky groups such as phenyl and benzyl are large enough to cause steric repulsions to widen the angle at the bridging Si-X-Si. For an easily deformed Si-O-Si bridge, the linear arrangement will be most favourable.

When X is a second or third row element the angle subtended at X can now be much smaller (less than 100°). The longer Si....Si distances may be accounted for by the 'close contact' Si....Si distance not being attained.

Other phenyl and benzyl analogues of Group IV elements have been investigated:

Table 8.4. R_3MXMR_3 compounds.

(where M is a Group IV element)

Compound	M - X(Å)	M-X-M(°)	Torsion angle(°)	Ref.
$(Ph_3C)_2O$	1.460(2)	127.9(1)	-	59
$(Ph_3Si)_2O$	1.616(1)	180.0(0)	60.0(0)	28
$(Bz_3Si)_2O$	1.613(4)	180.0(0)	60.0(0)	29
$(Ph_3Ge)_2O$	1.767(2)	1.352(2)	49.3(10)	60
$(Bz_3Ge)_2O$	1.730(1)	180.0(0)	60.0(0)	61
$(Ph_3Sn)_2O$	1.957(2)	137.3(1)	52.5(10)	62
$(Bz_3Sn)_2O$	1.919(0)	180.0(0)	60.0(0)	63

The mean torsion angle is defined as the absolute value of the mean angle between the projection of each R - M bond and the projection of the nearest R' - M' bond, onto a plane perpendicular to the line through M - M'.

From Table 8.4 it is seen that for the large phenyl and benzyl substituents, torsional angles are also large: the linear R_3MXMR_3 compounds have perfectly staggered conformations. The trend is towards those with phenyl groups being bent but with large M-X-M angles, whilst those with benzyl substituents are linear. Linear $Ph_3SiOSiPh_3$ is the exception, but as pointed out previously, the Si-O-Si skeleton shows somewhat unusual traits. Since the difference in bulkiness between benzyl and phenyl substituents will not be great, one can suggest that something other than steric factors is causing the benzyl analogues to be strictly linear. Tentatively, this may be attributed to electronic effects altering energy levels for the molecular orbitals in these benzyl analogues, so prohibiting distortion of the linear R_3MXMR_3 species.

Following this, obviously investigation of the structure of $[(PhCH_2)_3C]_2O$ would be worthwhile. Also of interest would be a benzyl analogue of a 'PPN' salt viz. $[(PhCH_2)_3PNP(CH_2Ph)_3]^+X^-$, and an investigation of the Si-S-Si angle in $Ph_3SiSSiPh_3$. Preparation of these latter two compounds was attempted, but proved to be unsuccessful.

Compounds of general formula $R_3SiNHSiR_3$ have just been mentioned. Many structures have also been investigated where the $R_3SiNSiR_3^-$ anion has been coordinated to a whole host of electrophiles. Table 8.5 shows that for R = Me, the Si-N-Si angle can be large, in some cases 136^o. The Si - N distance also varies substantially, but this is

unsurprising in view of the differing elecrophiles to which $R_3SiNSiR_3$ has been chelated. The longest Si - N bonds reported are in $TaCl_3[N(SiMe_3)_2]_2$, 1.785 - 1.826(7) Å. The N - Ta distances, 1.928(7) and 1.933(7) Å are very short, indicative of quite strong N ⟶ Ta π-bonding.

Table 8.5. Some $M[N\{Si(CH_3)_3\}_2]^-$ compounds[64].

Compound	Si - N(Å)	Si-N-Si(°)
$K[N\{SiMe_3\}_2].2C_4H_8O_2$(66)	1.64(1)	136.2(12)
$Na[N\{SiMe_3\}_2]$	1.690(5)	125.6(1)
$Be[N\{SiMe_3\}_2]_2$	1.722(7)	129.2(7)
$H[N\{SiMe_3\}_2]$ (49)	1.735(12)	125.5(18)
$Fe[N\{SiMe_3\}_2]_3$	1.731(3)	121.2 (planar complex)
$Al[N\{SiMe_3\}_2]_3$	1.75(2)	118.0(15)
$TaCl_3[N\{SiMe_3\}_2]_2$(65)	1.785 - 1.826(7)	

Sheldrick and Domingos found very short Si - N bonds in bis(dioxane)potassium bis(trimethylsilylamide). In this compound the N coordinates to the potassium ion, which is also bridged by the 4 oxygen atoms of the dioxane molecules. Si - N distances are only 1.64(1) Å[66].

Bond lengths and angles of the isolated $Me_3SiNSiMe_3^-$ anion would probably be interesting: however, interaction with the counter ion or solvent of crystallisation, to a greater or lesser degree, has been observed to date. One possible method may be to enclose the anion in a crown ether or cryptate, to isolate it from any cations or electron acceptors.

XI. Geometries of Molecular Pseudohalides.

The MNDO[55] work described in Chapter Seven was undertaken to try to rationalise the effect of variation of the R group in a pseudohalide molecule, RXYZ, with the calculated force constant for bending of the linear geometry in the plane of the molecule.

Table 7.2 is reproduced overleaf. Different R groups cause little variation in values of f_Q, and any set trends are difficult to recognise. However, trends in the geometries of various types of pseudohalide are obvious. Taking first those pseudohalides in which carbon is bonded to atom X:

(i) For all azides, f_Q is large and negative. Therefore all azides are bent.

Table 7.2(a). Force Constants for bending of linear geometries (Nm^{-1}).

	XYZ =			
R =	-NNN	-NCO	-CNO	-NCS
H-	-101.028	-28.079	13.208	-8.132
F-	* molecule dissociates	-79.175	-	-40.920
Cl-	*molecule dissociates	-32.575	0.206	-15.294
CH_3-	-71.336	-16.155	8.738	-2.273
CH_2F-	-92.485	-25.992	9.725	-7.679
CF_2H-	-102.679	-24.064	11.106	-4.381
CF_3-	-62.912	-13.257	12.576	-2.839
CH_2Cl-	-92.779	-27.919	4.872	-10.124
CCl_2H-	-89.965	-22.740	10.100	-4.815
Cl_3C-	-64.167	-13.065	12.962	-1.812
F_2ClC-	-61.484	-15.116	19.331	-6.117
Cl_2FC-	-61.371	-14.706	6.509	-2.000
HFClC-	-91.670	-22.000	5.393	-9.051

Table 7.2(b).

	-XYZ =		
R =	-NNN	-NCO	-NCS
H_3Si-	-12.566	1.946	4.259×10^{-3}
H_2FSi-	-19.833	-1.654	4.929×10^{-3}
F_2HSi-	-15.433	-2.036	4.895×10^{-3}
F_3Si-	-12.124	3.333	4.150×10^{-3}
H_2ClSi-	-24.423	-3.856	6.908×10^{-3}
Cl_2HSi-	-24.105	-3.582	6.857×10^{-3}
Cl_3Si-	-10.371	4.028	3.936×10^{-3}
F_2ClSi-	-15.845	8.077	8.226×10^{-3}
Cl_2FSi-	-21.935	1.930	4.961×10^{-3}
$HFClSi-$	-21.924	-1.036	2.743×10^{-3}

(ii) For all cyanates (-NCO), f_Q is negative, but takes a much smaller value than the analogous azides. These, too, are all bent molecules.

(iii) For thiocyanates, f_Q is quite small, but still a negative value. Again, these molecules will have bent optimum geometries.

(iv) For all fulminates (-CNO), f_Q values are positive so the preferred geometries of these molecules will be linear.

Considering, secondly, those pseudohalides in which silicon is bonded to atom X:

(i) For all silicon-containing azides, f_Q is again negative, but actual values of f_Q are much less than for the corresponding carbon pseudohalides. All silicon azides will nevertheless still be bent.

(ii) For silicon-containing cyanates, some f_Q values are positive, and some negative. All have rather small numerical values. Considering the angles γ for the optimised geometries, these range from 170 to 180°.

(iii) For silicon-containing thiocyanates, all calculated values of f_Q are essentially zero.

From these values, one can conclude that force constants for bending of molecular pseudohalides are much larger and more negative, that is bending of the molecule is more favoured, when R contains a carbon atom rather than silicon. Put another way, having a silicon atom in the R group will tend to help the pseudohalide to adopt a linear geometry.

Experimentally determined structures have been reported for several pseudohalides, and agreement with the MNDO work is excellent. Some of these pseudohalides are given in Table 8.6:

Table 8.6. Geometries of some Molecular Pseudohalides.

RXYZ	R-X-Y(°)	Ref.
HNNN	114.1	67
ClNNN	109	68
MeNNN	117	68
H_3SiNNN	124	68
H_3GeNNN	119	68
HNCO	128.1	67
MeNCO	140	69
H_3SiNCO	152	69
F_3SiNCO	161	69
Cl_3SiNCO	138	69
HNCS	135.0	67
H_3SiNCS	164	69
HCNO	180.0	67

Silyl isocyanate has also been examined by low temperature single-crystal X-ray work (140K)[70]. In the solid state, the Si-N-C angle is 158°. Electron diffraction work[69] also calculated that the molecule is bent, but also suggested that the energy required to change the Si-N-C angle from 140° to 180° is very low, only 100 to 250 cm^{-1}. From MNDO work (Table 7.2(b)), the force constant for bending the linear molecule is very small, only 1.946 Nm^{-1}., which is an example of the correlation between experimentally determined and

calculated geometries.

Chapter Eight.

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Appendix A.

Publications.

1.C.Glidewell, H.D.Holden, D.C.Liles. J. Chem. Res.(M), 4364-4372(1978).

'The Crystal and Molecular Structure of 4-N-(4'-bromophenylamino)-3-butylfuroxan.'

2. A.F.Cuthbertson, C.Glidewell, H.D.Holden, D.C.Liles. J. Chem. Res.(M), 3714-3733(1979).

'The Constitution of Dost's Keto Compound: Crystal and Molecular structure of 4-phenyl-3-phenylamino-1,2,4-thiadiazolin-5-one.'

3. C.Glidewell, H.D.Holden, D.C.Liles. Acta Cryst. B36, 1244-1247(1980).

'The Structure of 5,6-dihydro-3-thiobenzoylmethylene-3H-thiazolo[2,3-c][1,2,4]thiadiazole.'

4. C.Glidewell, H.D.Holden, D.C.Liles. J. Mol. Struct. 66, 325-331(1980).

'The Crystal and Molecular structure of 4,4'-methylenebis(1,3,5-trimethyl-4-imidazolin-2-one).'

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'The Structure of 1,1,1,3,3,3-hexaphenyldisilazane.'

6. C.Glidewell, G.S.Harris, H.D.Holden, D.C.Liles, J.S.McKechnie. J. Fluor. Chem. 18, 143-154(1981).
'Reactions of triphenylarsineoxide with aqueous hydrogen fluoride: crystal structure of bis(triphenylarsineoxide) hydrogen(I) tetrafluoroborate.'

7. C.Glidewell, H.D.Holden. In press.
'Hexa-organo substituted triatomics R_3XYZR_3: the configuration of $(R_3PNPR_3)^+$ and the crystal and molecular structure of triphenyl(P,P,P-triphenylphosphine imidato-N) phosphorus(I) thiocyanate.'

8. C.Glidewell, H.D.Holden. In press.
'The structure of bis(4-methylpyridine)hydrogen(I) tetraphenylborate.'

9. C.Glidewell, H.D.Holden. Submitted for publication.
'Factors Influencing the stability of cations $(Amine-H-Amine)^+$ and related ions.'

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Structural Studies of Some Heterocyclic and Organometallic Compounds.

being a thesis

presented by

Hazel Diane Holden, B.Sc.

to the

University of St. Andrews

in application for

the degree of Doctor of Philosophy.

St. Andrews September 1981

Supplementary Data.

I. 5,6-Dihydro-1-thiobenzoylmethylene-1H-thiazolo-[2,3-c][1,2,4]thiadiazole.

(i) Structure Factors

(ii) Thermal Parameters

OBSERVED AND CALCULATED STRUCTURE FACTORS

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
4	0	0	1122	-1082	25	3	0	236	257	-3	1	1	567	-558	-1	3	1	141	238	7	5	1	392	423
6	0	0	676	-695	0	4	0	29	-305	1	1	1	1252	-1280	1	3	1	735	-896	11	5	1	288	-297
8	0	0	1071	-1108	2	4	0	361	507	3	1	1	784	-811	3	3	1	373	-379	13	5	1	427	-447
10	0	0	380	389	4	4	0	146	-218	5	1	1	657	609	5	3	1	461	-457	15	5	1	224	-237
14	0	0	651	-669	6	4	0	897	-901	7	1	1	212	-213	7	3	1	780	-802	-20	6	1	258	-212
16	0	0	147	220	12	4	0	993	917	11	1	1	668	-677	9	3	1	719	771	-16	6	1	115	-112
18	0	0	981	-913	14	4	0	696	696	15	1	1	432	472	11	3	1	235	-201	-14	6	1	185	-193
22	0	0	155	168	16	4	0	315	-373	17	1	1	455	401	13	3	1	502	489	-12	6	1	365	285
26	0	0	497	589	20	4	0	308	-306	19	1	1	576	559	15	3	1	501	573	-10	6	1	204	214
3	1	0	566	-552	22	4	0	178	-231	23	1	1	179	-186	25	3	1	202	252	-8	6	1	75	77
7	1	0	173	166	1	5	0	141	245	27	1	1	185	-252	-22	4	1	190	-191	-4	6	1	335	-400
9	1	0	459	473	3	5	0	189	-239	-26	2	1	179	229	-16	4	1	271	331	-2	6	1	184	234
11	1	0	251	-231	5	5	0	288	325	-24	2	1	291	340	-10	4	1	90	-107	0	6	1	193	-348
13	1	0	896	901	13	5	0	585	-562	-20	2	1	455	465	-8	4	1	700	740	2	6	1	178	-224
17	1	0	449	416	17	5	0	227	186	-18	2	1	525	-479	-6	4	1	279	277	4	6	1	198	215
21	1	0	210	-221	2	6	0	48	-99	-12	2	1	1517	-1353	-2	4	1	208	311	6	6	1	205	-249
2	2	0	1785	-2045	4	6	0	106	145	-10	2	1	320	-351	0	4	1	152	-223	10	6	1	110	116
4	2	0	168	137	6	6	0	273	301	-8	2	1	114	-142	2	4	1	429	-505	14	6	1	519	525
6	2	0	688	658	10	6	0	159	-135	-6	2	1	1265	-1251	4	4	1	513	-523	16	6	1	170	188
8	2	0	1185	1219	12	6	0	317	-297	-4	2	1	493	521	6	4	1	720	-653	20	6	1	252	216
10	2	0	183	-133	1	7	0	39	-73	-2	2	1	1760	-1884	12	4	1	161	-134	-15	7	1	277	273
12	2	0	792	-743	3	7	0	83	-135	0	2	1	1393	1623	16	4	1	132	-172	-13	7	1	131	-123
14	2	0	380	-405	5	7	0	251	-293	2	2	1	2825	3011	20	4	1	190	170	-11	7	1	169	-172
16	2	0	291	337	9	7	0	196	196	6	2	1	540	538	-21	5	1	203	210	-7	7	1	223	-231
18	2	0	781	725	13	7	0	293	244	8	2	1	682	614	-17	5	1	226	-284	1	7	1	146	210
20	2	0	214	221	0	8	0	27	316	10	2	1	211	206	-15	5	1	114	-129	5	7	1	169	-190
22	2	0	171	203	12	8	0	232	-215	14	2	1	926	-1022	-13	5	1	273	-212	7	7	1	158	148
3	3	0	276	-293	7	9	0	91	66	18	2	1	164	148	-11	5	1	344	-357	9	7	1	137	-141
5	3	0	260	285	11	9	0	144	-178	20	2	1	608	-574	-9	5	1	165	138	11	7	1	254	199
7	3	0	289	-267	-17	1	1	260	-250	-19	3	1	350	-312	-7	5	1	195	-199	-12	8	1	126	-99
9	3	0	202	-189	-15	1	1	413	-411	-17	3	1	471	380	-5	5	1	156	-165	-6	8	1	91	90
11	3	0	340	-348	-13	1	1	487	403	-15	3	1	142	150	-3	5	1	58	85	2	8	1	63	88
13	3	0	458	-419	-11	1	1	807	887	-13	3	1	579	546	-1	5	1	115	-212	-11	9	1	97	88
15	3	0	182	-191	-9	1	1	661	700	-9	3	1	542	-499	1	5	1	232	365	-9	9	1	81	109
17	3	0	155	-162	-7	1	1	1260	1237	-7	3	1	274	238	3	5	1	470	554	-7	9	1	161	193
19	3	0	227	-214	-5	1	1	258	-261	-3	3	1	575	586	5	5	1	373	446	-3	9	1	63	-105

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
1	9	1	62	-120	11	1	2	198	189	11	3	2	149	-115	-22	6	2	235	-181	-11	1	3	107	50
5	9	1	103	-152	13	1	2	278	280	13	3	2	155	-126	-16	6	2	202	204	-9	1	3	585	-614
10	10	1	114	-130	15	1	2	282	-314	15	3	2	518	566	-12	6	2	109	100	-7	1	3	660	621
-26	0	2	171	219	17	1	2	567	-528	21	3	2	271	276	-10	6	2	132	-157	-5	1	3	421	406
-20	0	2	544	-568	-18	2	2	496	408	-24	4	2	267	274	-8	6	2	78	-85	-3	1	3	1472	-1416
-18	0	2	893	-811	-16	2	2	264	295	-16	4	2	476	-500	-6	6	2	224	247	-1	1	3	220	214
-16	0	2	388	-415	-14	2	2	282	275	-14	4	2	204	184	-4	6	2	160	226	1	1	3	830	-861
-14	0	2	808	-848	-12	2	2	824	-734	-12	4	2	184	210	-2	6	2	200	256	5	1	3	619	528
-12	0	2	1079	996	-10	2	2	698	-655	-10	4	2	230	-228	0	6	2	60	-78	7	1	3	772	795
-10	0	2	3044	3138	-8	2	2	782	-793	-8	4	2	194	195	2	6	2	278	-354	9	1	3	881	855
-8	0	2	171	-137	-6	2	2	145	-138	-6	4	2	797	-845	4	6	2	339	-373	11	1	3	176	198
-6	0	2	906	947	-4	2	2	716	700	-4	4	2	475	-523	6	6	2	141	-166	15	1	3	186	-183
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3	5	8	272	-314	0	2	9	620	681	-2	8	9	143	176	-7	3	10	508	467	-4	2	11	707	665
5	5	8	164	171	2	2	9	299	221	1	9	9	133	-114	-5	3	10	309	271	4	2	11	308	-303
-14	6	8	215	-170	4	2	9	412	359	-20	0	10	354	-404	-3	3	10	213	-170	8	2	11	471	-468
-8	6	8	223	239	6	2	9	370	369	-18	0	10	494	-553	3	3	10	159	150	-17	3	11	166	216
-4	6	8	106	109	12	2	9	296	-248	-14	0	10	604	573	7	3	10	573	-554	-15	3	11	202	215
-2	6	8	110	-82	14	2	9	403	-463	-12	0	10	1062	1051	15	3	10	205	183	-11	3	11	513	-493
0	6	8	161	-183	-23	3	9	159	-182	-10	0	10	571	543	-24	4	10	227	236	-9	3	11	592	-535
4	6	8	171	196	-19	3	9	261	257	-8	0	10	217	-191	-16	4	10	224	-219	-5	3	11	176	-120
6	6	8	122	117	-17	3	9	348	315	-6	0	10	577	489	-14	4	10	214	189	3	3	11	368	362
8	6	8	256	-209	-11	3	9	745	-608	-4	0	10	590	-583	-6	4	10	908	-738	5	3	11	169	-172
0	6	8	267	-199	-9	3	9	312	-325	0	0	10	188	169	-4	4	10	206	-186	9	3	11	352	370
3	7	8	138	158	-7	3	9	329	381	4	0	10	171	-134	0	4	10	545	447	-8	4	11	228	-216
5	7	8	190	-196	-5	3	9	249	280	6	0	10	1117	-1070	2	4	10	749	638	6	4	11	216	179
1	7	8	221	-173	-3	3	9	193	185	8	0	10	691	-725	-17	5	10	173	142	-15	5	11	198	-190
7	7	8	153	125	-1	3	9	393	-425	12	0	10	171	176	-9	5	10	221	-192	-13	5	11	250	-231
-6	8	8	120	116	1	3	9	716	-783	14	0	10	424	426	-5	5	10	288	253	-11	5	11	385	364
-4	8	8	145	131	9	3	9	498	445	16	0	10	242	305	1	5	10	542	-444	-9	5	11	463	387
-2	8	8	189	154	13	3	9	238	269	-19	1	10	282	352	-6	6	10	274	211	-7	5	11	316	267
-17	1	9	337	-313	-4	4	9	232	-240	-17	1	10	173	-152	-4	6	10	249	205	-5	5	11	396	365
-13	1	9	1560	1468	2	4	9	198	-210	-15	1	10	342	320	-2	6	10	240	-202	3	5	11	243	-249
-11	1	9	961	811	12	4	9	186	186	-11	1	10	761	-723	0	6	10	227	-198	7	5	11	221	-194
-9	1	9	423	484	-17	5	9	268	-272	3	1	10	353	316	-7	7	10	205	-130	9	5	11	273	-265
-5	1	9	706	-791	-13	5	9	279	-215	15	1	10	219	-274	1	7	10	440	310	-6	6	11	301	-228
-3	1	9	184	-202	-11	5	9	200	142	-18	2	10	190	187	3	7	10	279	206	8	6	11	244	194
1	1	9	272	-331	-7	5	9	225	-249	-12	2	10	589	-549	7	7	10	186	130	-7	7	11	267	-211
3	1	9	422	370	-1	5	9	314	327	-10	2	10	438	-424	-6	8	10	142	143	3	7	11	202	-118
5	1	9	540	-532	1	5	9	335	331	-6	2	10	615	558	6	8	10	193	-149	-22	0	12	255	320
7	1	9	683	-652	5	5	9	315	278	-4	2	10	320	310	-19	1	11	357	418	-16	0	12	436	478
13	1	9	425	419	9	5	9	338	-319	0	2	10	659	-580	-13	1	11	302	351	-14	0	12	284	-286
-16	2	9	271	-250	13	5	9	223	-242	2	2	10	344	-308	-9	1	11	232	222	-10	0	12	196	203
-14	2	9	510	-481	-14	6	9	195	115	4	2	10	404	341	-7	1	11	216	184	-4	0	12	1016	-1039
-12	2	9	538	-478	-8	6	9	303	328	8	2	10	379	392	-5	1	11	567	-586	-2	0	12	793	-832
-8	2	9	381	-399	-4	6	9	187	-196	-19	3	10	266	-253	-1	1	11	374	-362	4	0	12	383	379
-6	2	9	184	-146	0	6	9	344	-333	-15	3	10	230	-231	3	1	11	485	485	10	0	12	199	222

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
-13	1	12	374	-410	8	4	12	246	-253	10	2	13	233	260	-12	2	14	280	266	-2	2	15	201	-253
-9	1	12	212	221	10	4	12	404	-393	-17	3	13	294	-313	-8	2	14	169	-155	0	2	15	206	242
1	1	12	329	355	-17	5	12	228	257	-15	3	13	213	-184	-6	2	14	268	-287	6	2	15	209	210
3	1	12	427	-478	-9	5	12	369	-342	-5	3	13	365	-388	0	2	14	181	177	-17	3	15	202	-217
5	1	12	155	-156	1	5	12	221	-201	-1	3	13	355	350	6	2	14	188	-228	-15	3	15	289	-356
-16	2	12	331	340	9	5	12	217	230	1	3	13	334	311	-15	3	14	199	-217	-9	3	15	323	382
-10	2	12	310	-327	11	5	12	227	243	-6	4	13	174	165	3	3	14	378	424	-7	3	15	193	208
-2	2	12	337	345	-8	6	12	239	-172	-17	5	13	195	226	-10	4	14	246	246	-9	5	15	288	-304
8	2	12	165	144	-9	7	12	324	239	1	5	13	373	-360	-8	4	14	246	218	-7	5	15	214	-240
-13	3	12	200	231	-7	7	12	238	168	-2	6	13	257	233	-4	4	14	239	244	-14	0	16	181	-240
-5	3	12	330	-302	1	7	12	256	223	-22	0	14	236	302	0	4	14	348	-362	-10	0	16	175	-207
-1	3	12	197	-170	9	7	12	245	-199	-18	0	14	202	286	-9	5	14	344	-357	-6	0	16	391	468
3	3	12	296	253	-17	1	13	154	176	-14	0	14	530	-611	-8	6	14	185	-149	-4	0	16	302	340
-18	4	12	179	-198	-13	1	13	174	-199	-12	0	14	316	-345	-9	7	14	326	299	-2	0	16	360	416
-16	4	12	412	-405	-11	1	13	400	-423	-10	0	14	290	-334	-15	1	15	283	321	-7	3	16	317	390
-14	4	12	218	-232	-3	1	13	273	241	4	0	14	557	648	-13	1	15	228	244	-15	1	17	199	271
-12	4	12	152	-147	5	1	13	318	-347	12	0	14	243	-291	-3	1	15	287	307	-13	1	17	309	423
-10	4	12	309	297	-16	2	13	324	359	-9	1	14	182	211	1	1	15	161	-146	-7	1	17	297	-377
-8	4	12	391	333	-14	2	13	325	329	1	1	14	255	-317	3	1	15	294	-373	-5	1	17	334	-406
-4	4	12	214	171	-10	2	13	219	257	13	1	14	165	236	5	1	15	357	-424	-14	2	17	187	-255
0	4	12	162	148	-4	2	13	207	-198	-14	2	14	258	258	-8	2	15	276	-314	0	2	17	179	202
2	4	12	343	351																				

Table A. Anistropic Thermal Parameters. ($/\text{Å}^2 \times 10^3$)

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{31}	U_{12}
S(1)	46(2)	69(2)	28(1)	2(2)	9(1)	-4(2)
S(2)	37(2)	86(3)	48(2)	5(2)	6(1)	7(2)
S(3)	57(2)	87(3)	37(2)	11(2)	20(1)	7(2)
N(1)	42(6)	78(8)	31(5)	-1(5)	-1(4)	3(5)
N(2)	36(5)	55(6)	22(4)	5(4)	7(4)	6(4)
C(1)	34(6)	46(7)	30(5)	-9(5)	10(5)	-2(5)
C(2)	35(6)	82(9)	26(6)	1(7)	2(5)	9(7)
C(3)	52(8)	108(13)	88(11)	39(7)	38(8)	31(8)
C(4)	43(7)	86(9)	32(6)	10(7)	21(5)	7(7)
C(5)	34(6)	65(8)	21(5)	0(5)	8(5)	9(6)
C(6)	40(6)	47(7)	35(6)	-6(5)	11(5)	-1(5)
C(7)	31(6)	45(6)	27(5)	-10(5)	5(4)	1(5)
C(8)	39(6)	52(7)	30(6)	0(5)	9(5)	2(6)
C(9)	54(8)	51(8)	42(7)	8(6)	14(6)	16(6)
C(10)	37(7)	68(9)	48(8)	-16(7)	5(6)	6(7)
C(11)	36(7)	58(8)	61(8)	-8(7)	13(6)	-1(6)
C(12)	37(6)	55(8)	56(8)	1(6)	18(6)	-3(6)

The anisotropic temperature factor takes the form

$$\exp[-2\pi^2(h^2\underline{a}^{*2}\underline{U}_{11} + k^2\underline{b}^{*2}\underline{U}_{22} + l^2\underline{c}^{*2}\underline{U}_{33} + 2kl\underline{b}^{*}\underline{c}^{*}\underline{U}_{23} + 2lh\underline{c}^{*}\underline{a}^{*}\underline{U}_{31} + 2hk\underline{a}^{*}\underline{b}^{*}\underline{U}_{12})]$$

The isotropic temperature factor for the unique hydrogen atom, H(5), refined to $15(6) \times 10^{-2}$ Å^2, the common isotropic temperature factor for H(31), H(32), H(41) and H(42) refined to $14(3) \times 10^{-2}$ Å^2, and that for the five phenyl hydrogens to $56(15) \times 10^{-3}$ Å^2.

II. 4-N-(4′-bromophenylamino)-3-butylfuroxan.

(i) Structure Factors.

(ii) Thermal Parameters.

OBSERVED AND CALCULATED STRUCTURE FACTORS FOR 4-N-(4'-BROMOPHENYL)AMINO-3-BUTYL FUROXAN

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
24	0	0	343	-369	9	1	0	190	185	17	3	0	115	-85	12	4	0	132	125	2	6	0	444	-421
22	0	0	242	-202	11	1	0	463	416	15	3	0	88	81	14	4	0	273	-259	0	6	0	231	139
20	0	0	199	-161	13	1	0	178	-175	13	3	0	722	-699	18	4	0	141	-133	2	6	0	431	-421
18	0	0	225	-264	15	1	0	208	201	9	3	0	445	-456	20	4	0	131	-157	4	6	0	604	-643
16	0	0	202	178	19	1	0	116	-111	7	3	0	771	-780	22	4	0	130	-91	6	6	0	269	-299
14	0	0	158	182	21	1	0	137	61	5	3	0	757	-697	24	4	0	166	-149	8	6	0	86	-136
12	0	0	901	882	23	1	0	170	-184	3	3	0	1016	-993	25	5	0	121	-131	10	6	0	276	-235
10	0	0	393	411	28	2	0	150	118	1	3	0	1550	-1480	23	5	0	99	-46	12	6	0	255	282
8	0	0	939	974	24	2	0	110	95	1	3	0	1550	-1480	21	5	0	166	-118	14	6	0	93	-105
6	0	0	1176	1249	22	2	0	120	147	3	3	0	1000	-993	17	5	0	147	129	16	6	0	307	288
4	0	0	1466	1556	20	2	0	144	180	5	3	0	739	-697	15	5	0	290	-244	20	6	0	95	136
4	0	0	1428	1556	18	2	0	92	98	7	3	0	755	-780	13	5	0	396	398	24	6	0	99	46
6	0	0	1146	1249	14	2	0	279	294	9	3	0	431	-456	11	5	0	415	376	21	7	0	116	148
8	0	0	916	974	12	2	0	537	-553	13	3	0	712	-699	9	5	0	622	582	19	7	0	112	-76
10	0	0	383	411	10	2	0	665	-600	15	3	0	112	81	7	5	0	894	936	15	7	0	98	96
12	0	0	878	882	8	2	0	1527	-1434	17	3	0	92	-85	5	5	0	762	762	13	7	0	152	-150
14	0	0	154	182	6	2	0	1260	-1157	19	3	0	123	64	3	5	0	593	590	11	7	0	254	-286
16	0	0	206	178	4	2	0	2925	-3049	21	3	0	100	50	1	5	0	1841	1828	9	7	0	389	-375
18	0	0	229	-264	2	2	0	1153	-1116	27	3	0	140	115	1	5	0	1833	1828	7	7	0	924	-927
20	0	0	193	-161	0	2	0	1726	-1994	26	4	0	141	-56	3	5	0	587	590	5	7	0	319	-323
22	0	0	207	-202	2	2	0	1153	-1116	24	4	0	167	-149	5	5	0	752	762	3	7	0	1283	-1280
24	0	0	333	-369	4	2	0	2917	-3049	20	4	0	156	-157	7	5	0	886	936	1	7	0	538	-514
27	1	0	128	3	6	2	0	1222	-1157	18	4	0	108	-133	9	5	0	604	582	1	7	0	541	-514
23	1	0	180	-184	8	2	0	1497	-1434	14	4	0	273	-259	11	5	0	404	376	3	7	0	1268	-1280
21	1	0	133	61	10	2	0	648	-600	12	4	0	141	125	13	5	0	391	398	5	7	0	300	-323
19	1	0	121	-111	12	2	0	526	-553	10	4	0	736	748	15	5	0	270	-244	7	7	0	909	-927
15	1	0	206	201	14	2	0	259	294	8	4	0	334	347	17	5	0	142	129	9	7	0	393	-375
13	1	0	183	-175	16	2	0	92	40	6	4	0	1000	985	19	5	0	96	-16	11	7	0	244	-286
11	1	0	437	416	18	2	0	98	98	4	4	0	692	669	21	5	0	144	-118	13	7	0	143	-150
9	1	0	202	185	20	2	0	159	180	2	4	0	1275	1225	20	6	0	130	136	17	7	0	106	107
7	1	0	97	-73	22	2	0	129	147	0	4	0	2016	1953	16	6	0	296	288	21	7	0	134	148
5	1	0	417	367	24	2	0	124	95	2	4	0	1268	1225	12	6	0	266	282	25	7	0	109	59
3	1	0	245	-291	26	2	0	148	139	4	4	0	675	669	10	6	0	273	-235	24	8	0	140	-80
3	1	0	241	-291	28	2	0	141	119	6	4	0	970	985	8	6	0	82	-136	14	8	0	112	-101
5	1	0	404	367	27	3	0	112	115	8	4	0	327	347	6	6	0	294	-299	12	8	0	265	-265
7	1	0	95	-73	19	3	0	94	64	10	4	0	719	748	4	6	0	690	-643	10	8	0	92	104

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
8	8	0	152	-169	10	10	0	453	475	-19	1	1	586	607	10	2	1	79	57	-18	4	1	240	-226
6	8	0	125	-78	14	10	0	166	-145	-17	1	1	649	606	12	2	1	473	-436	-16	4	1	366	-332
4	8	0	142	132	13	11	0	199	-178	-15	1	1	598	584	14	2	1	746	-761	-14	4	1	840	-807
2	8	0	93	-65	11	11	0	203	222	-13	1	1	1077	1082	16	2	1	115	-91	-12	4	1	616	-582
0	8	0	73	20	9	11	0	129	-154	-11	1	1	1191	1266	18	2	1	351	-303	-10	4	1	120	-106
2	8	0	89	-65	7	11	0	147	-156	-9	1	1	131	171	20	2	1	147	-170	-8	4	1	810	-838
4	8	0	136	132	3	11	0	346	-359	-7	1	1	2375	2379	26	2	1	114	-87	-6	4	1	192	-206
6	8	0	114	-78	1	11	0	176	-188	-5	1	1	1084	1097	-23	3	1	131	-143	-4	4	1	878	-785
8	8	0	152	-169	1	11	0	186	-188	-3	1	1	245	272	-21	3	1	201	-134	-2	4	1	655	601
12	8	0	267	-265	3	11	0	321	-359	1	1	1	1764	-1972	-19	3	1	316	-300	0	4	1	924	-921
14	8	0	89	-101	5	11	0	100	54	3	1	1	231	-197	-17	3	1	267	-347	2	4	1	771	800
18	8	0	95	-117	7	11	0	133	-158	5	1	1	1275	-1266	-15	3	1	970	-945	4	4	1	97	151
20	8	0	118	-46	9	11	0	138	-154	7	1	1	1550	-1478	-13	3	1	646	-646	6	4	1	372	329
23	9	0	111	-56	11	11	0	195	222	9	1	1	863	-903	-11	3	1	832	-818	8	4	1	1146	1036
13	9	0	174	-171	13	11	0	215	-178	11	1	1	1214	-1226	-9	3	1	616	-563	10	4	1	817	869
11	9	0	286	301	16	12	0	105	-23	13	1	1	1191	-1163	-7	3	1	1283	-1140	12	4	1	265	308
9	9	0	325	320	14	12	0	117	-72	15	1	1	594	-602	-5	3	1	160	-167	14	4	1	531	486
7	9	0	212	185	10	12	0	108	-131	17	1	1	748	-772	-3	3	1	730	-690	16	4	1	287	278
5	9	0	341	348	6	12	0	186	-185	19	1	1	346	-346	-1	3	1	68	-22	18	4	1	371	386
3	9	0	536	540	4	12	0	118	85	21	1	1	349	-325	1	3	1	1695	1452	20	4	1	280	264
1	9	0	419	447	2	12	0	257	-243	-24	2	1	126	110	3	3	1	226	-264	22	4	1	164	95
1	9	0	419	447	0	12	0	95	-119	-20	2	1	305	275	5	3	1	631	643	24	4	1	122	104
3	9	0	528	540	2	12	0	238	-243	-18	2	1	262	264	7	3	1	1039	1145	-19	5	1	118	116
5	9	0	338	348	4	12	0	102	85	-16	2	1	199	207	9	3	1	1634	1524	-17	5	1	325	307
7	9	0	199	185	6	12	0	188	-185	-14	2	1	88	-70	11	3	1	735	706	-15	5	1	318	304
9	9	0	324	320	10	12	0	110	-131	-12	2	1	350	373	13	3	1	787	844	-13	5	1	451	437
11	9	0	299	301	11	13	0	112	-20	-10	2	1	469	426	15	3	1	379	399	-11	5	1	503	461
13	9	0	177	-171	7	13	0	129	69	-8	2	1	387	374	17	3	1	412	382	-9	5	1	367	387
23	9	0	152	-56	3	13	0	105	147	-6	2	1	1245	1163	19	3	1	342	328	-7	5	1	325	345
14	10	0	150	-145	5	13	0	105	-29	-4	2	1	651	604	21	3	1	128	121	-5	5	1	121	92
10	10	0	470	475	2	14	0	121	100	-2	2	1	457	367	23	3	1	157	157	-3	5	1	100	-33
8	10	0	164	-154	2	14	0	137	100	0	2	1	2039	-1830	-28	4	1	120	-12	-1	5	1	243	278
6	10	0	369	359	10	14	0	150	125	2	2	1	497	-401	-26	4	1	106	-73	1	5	1	71	-49
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17	5	1	234	-239	19	7	1	167	-165	-18	10	1	155	136	12	12	1	117	42	-11	1	2	120	134
-24	6	1	112	128	-22	8	1	141	-94	-14	10	1	107	128	-5	13	1	109	-99	-9	1	2	1077	-784
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-18	6	1	344	335	-16	8	1	415	-403	-10	10	1	376	367	-1	13	1	102	50	-5	1	2	2192	2174
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-17	7	1	93	-47	-19	9	1	108	130	11	11	1	250	-240	4	0	2	1711	1759	-6	2	2	1871	-1848
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22	2	2	276	294	10	4	2	99	91	2	6	2	209	-213	2	8	2	273	-263	-7	11	2	199	-189
24	2	2	104	92	14	4	2	158	-170	4	6	2	625	-600	4	8	2	135	116	-5	11	2	105	-106
26	2	2	141	123	16	4	2	105	-118	6	6	2	311	321	6	8	2	312	-310	-3	11	2	361	-391
-25	3	2	99	57	18	4	2	299	-305	8	6	2	203	-198	8	8	2	268	-261	1	11	2	157	-186
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9	3	2	108	-144	5	5	2	787	740	-1	7	2	414	-414	5	9	2	563	584	14	13	2	153	-90
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-9	1	3	886	797	20	2	3	160	-150	2	4	3	771	741	-14	6	3	477	487	-8	8	3	181	-191
-7	1	3	665	743	24	2	3	117	-92	4	4	3	603	564	-12	6	3	189	176	-6	8	3	244	-238
-5	1	3	886	-858	-21	3	3	276	-306	6	4	3	231	242	-10	6	3	735	726	-4	8	3	182	-142
-3	1	3	477	438	-19	3	3	254	-269	8	4	3	676	696	-6	6	3	377	383	-2	8	3	96	96
-1	1	3	147	-110	-17	3	3	425	-417	10	4	3	232	194	-4	6	3	86	-134	0	8	3	109	-100
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9	1	3	810	-857	-7	3	3	459	-455	20	4	3	207	193	8	6	3	894	-910	10	8	3	317	319
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8	2	3	825	-816	-8	4	3	150	-101	-24	6	3	100	86	-18	8	3	217	-196	-10	10	3	104	141
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2	10	3	176	-197	-16	0	4	420	428	-18	2	4	79	-33	-20	4	4	91	-7	19	5	4	116	-120
4	10	3	108	-105	-14	0	4	691	-753	-16	2	4	195	-220	-18	4	4	92	104	21	5	4	131	-81
6	10	3	254	-275	-12	0	4	2192	2216	-14	2	4	199	-189	-14	4	4	167	200	-18	6	4	96	-11
8	10	3	167	-195	-10	0	4	643	692	-12	2	4	661	-634	-12	4	4	417	383	-16	6	4	162	-164
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12	10	3	166	-139	-6	0	4	939	785	-8	2	4	1184	-1205	-8	4	4	421	414	-10	6	4	95	-24
14	10	3	160	-163	-4	0	4	1146	1083	-6	2	4	924	-1008	-6	4	4	738	736	-8	6	4	471	-467
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-9	11	3	93	16	2	0	4	764	773	0	2	4	1298	-1220	0	4	4	962	995	-2	6	4	539	-514
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-15	7	5	99	108	7	9	5	134	114	-8	0	6	328	335	2	2	6	392	-410	12	4	6	182	-183
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-2	12	7	129	37	-15	3	8	102	-98	6	6	8	181	169	11	1	9	114	-83	-13	7	9	110	99
-26	0	8	157	-159	-11	3	8	129	-185	10	6	8	143	142	-14	2	9	99	91	-9	7	9	113	-84
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-16	0	8	128	154	-7	3	8	182	-160	-11	7	8	116	-70	-2	2	9	163	-173	-3	7	9	113	54
-12	0	8	206	193	-5	3	8	121	-117	-9	7	8	186	-190	0	2	9	102	125	-1	7	9	116	91
-10	0	8	140	174	-3	3	8	209	-211	-5	7	8	247	-261	6	2	9	101	-27	-10	8	9	109	-58
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1	1	8	119	-98	0	4	8	95	79	-5	9	8	113	138	6	4	9	146	144	-11	1	10	110	-59
5	1	8	195	192	2	4	8	93	-91	-3	9	8	141	61	8	4	9	132	97	-9	1	10	99	89
13	1	8	118	14	6	4	8	95	-43	-1	9	8	111	125	-19	5	9	121	35	-7	1	10	93	71
-20	2	8	111	-71	10	4	8	128	-86	-12	10	8	143	112	-15	5	9	102	32	-1	1	10	129	120
-16	2	8	91	-59	12	4	8	169	-175	0	10	8	111	98	-5	5	9	94	-72	-18	2	10	113	-23
-14	2	8	111	-103	-11	5	8	272	277	2	10	8	134	102	-3	5	9	116	-97	-10	2	10	121	-111
-12	2	8	215	-197	-7	5	8	160	142	-9	11	8	108	-87	1	5	9	146	-135	-4	2	10	215	-188
-10	2	8	187	-192	-5	5	8	276	293	-13	1	9	141	126	3	5	9	153	-78	-2	2	10	97	50
-8	2	8	135	-165	-3	5	8	118	116	-11	1	9	102	-70	5	5	9	112	-111	2	2	10	112	-6
-6	2	8	214	-225	-1	5	8	278	306	-9	1	9	94	61	9	5	9	112	-56	-17	3	10	108	-40
-4	2	8	303	-286	3	5	8	107	39	-7	1	9	116	-95	-12	6	9	127	99	-9	3	10	112	-78
0	2	8	115	-136	5	5	8	254	243	-5	1	9	102	-28	-10	6	9	116	-78	-5	3	10	168	-144
2	2	8	301	-314	-22	6	8	117	49	-3	1	9	277	-263	-6	6	9	106	108	5	3	10	115	-38
4	2	8	151	173	-14	6	8	123	-97	-1	1	9	119	-90	-4	6	9	186	-176	-14	4	10	103	11
6	2	8	105	-70	-6	6	8	115	15	3	1	9	225	-212	0	6	9	118	-114	-6	4	10	126	107
10	2	8	115	69	0	6	8	113	123	5	1	9	98	-96	2	6	9	108	-75	-4	4	10	108	-65
12	2	8	127	88	2	6	8	108	-124	7	1	9	130	-171	6	6	9	148	-114	4	4	10	115	-6

H	K	L	10FO	10FC
-11	5	10	126	103
-7	5	10	121	133
-5	5	10	123	35
-3	5	10	115	-15
-1	5	10	114	139
-14	6	10	105	-36
-8	6	10	110	-31
-6	6	10	125	63
-4	6	10	153	-97
-2	6	10	128	22
0	6	10	127	79
-11	7	10	115	-85
-5	7	10	157	-98
-3	7	10	115	-55
-9	1	11	108	-87
-8	2	11	139	-92
-4	2	11	116	-4

Table 3 - Anisotropic thermal paramoters ($/Å^2 \times 10^3$)

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{31}	U_{12}
Br	90.8(8)	117.7(9)	109.8(9)	42.7(7)	-0.1(6)	20.7(6)
O(1)	100(4)	37(3)	87(4)	-5(3)	7(3)	2(3)
O(2)	125(5)	42(3)	100(5)	22(3)	-7(4)	17(3)
N(1)	79(4)	49(4)	69(4)	-9(3)	11(4)	-2(3)
N(2)	81(4)	40(4)	67(4)	0(3)	12(3)	-1(3)
N(3)	71(4)	41(3)	52(4)	3(3)	8(4)	-4(3)
C(1)	54(4)	45(4)	58(5)	-2(4)	22(4)	-1(3)
C(2)	65(4)	37(4)	46(4)	0(3)	16(4)	-1(3)
C(7)	73(5)	45(4)	59(5)	11(4)	23(4)	2(4)
C(8)	84(6)	55(5)	75(6)	7(5)	18(5)	12(4)
C(9)	93(6)	53(5)	87(6)	16(5)	21(5)	16(5)
C(10)	56(5)	73(6)	74(6)	16(5)	5(4)	7(4)
C(11)	67(5)	82(7)	84(6)	22(5)	2(5)	-1(5)
C(12)	78(5)	60(5)	66(5)	-5(5)	3(4)	-10(4)

The anisotropic temperature factor takes the form

$$\exp[-2\pi^2(h^2\underline{a}^{*2}\underline{U}_{11} + k^2\underline{b}^{*2}\underline{U}_{22} + l^2\underline{c}^{*2}U_{33} + 2kl\underline{b}^*\underline{c}^*\underline{U}_{23} + 2lh\underline{c}^*\underline{a}^*\underline{U}_{31} + 2hka^*\underline{b}^*\underline{U}_{12})]$$

The isotropic temperature factors refined to the following values:

C(3)	64(2)	x	10^{-3} Å^2
C(6)	147(4)	x	10^{-3} Å^2
H(10)	9(3)	x	10^{-2} Å^2

The common isotropic temperature factor for H(11), H(12), H(13) and H(14) refined to 13(2) x 10^{-2} Å^2: that for C(41), C(42), C(51) and C(52) refined to 100(3) x 10^{-3} Å^2.

III. 4,4′-Methylenebis(1,3,5-trimethyl-4-imidazoline-2-one.

(i) Structure Factors.

(ii) Thermal Parameters.

H	K	L	10FO	10FC
3	-13	0	36	50
7	-13	0	28	23
0	-12	0	3	41
1	-12	0	13	27
3	-12	0	31	40
4	-12	0	28	23
5	-12	0	31	23
8	-12	0	31	28
1	-11	0	16	20
2	-11	0	30	47
3	-11	0	24	33
4	-11	0	23	26
5	-11	0	38	41
6	-11	0	34	39
9	-11	0	30	37
0	-10	0	10	122
1	-10	0	11	18
2	-10	0	11	10
3	-10	0	33	34
5	-10	0	24	12
8	-10	0	54	50
10	-10	0	38	32
11	-10	0	64	56
2	-9	0	45	56
3	-9	0	137	150
4	-9	0	43	38
5	-9	0	67	63
6	-9	0	64	54
8	-9	0	44	36
9	-9	0	39	32
1	-8	0	63	95
2	-8	0	22	40
4	-8	0	95	102
5	-8	0	19	21
6	-8	0	44	53
7	-8	0	66	59
1	-7	0	19	4[illegible]
3	-7	0	19	29
4	-7	0	30	32
5	-7	0	119	103
6	-7	0	95	86
7	-7	0	43	42
8	-7	0	42	45
9	-7	0	72	59
10	-7	0	41	49
13	-7	0	46	50
0	-6	0	19	142
1	-6	0	23	26
2	-6	0	123	136
3	-6	0	103	132
4	-6	0	62	61
5	-6	0	52	52
7	-6	0	167	155
8	-6	0	105	127
10	-6	0	66	70
1	-5	0	34	35
2	-5	0	220	216
3	-5	0	47	55
4	-5	0	36	47
6	-5	0	134	127
7	-5	0	34	37
8	-5	0	107	135
9	-5	0	46	52
11	-5	0	39	39
0	-4	0	383	279
1	-4	0	63	112
2	-4	0	305	301
3	-4	0	85	86
4	-4	0	135	112
5	-4	0	145	102
6	-4	0	317	296
7	-4	0	98	103
8	-4	0	101	131
10	-4	0	83	82
12	-4	0	34	32
14	-4	0	36	56
1	-3	0	116	129
2	-3	0	67	62
3	-3	0	278	262
4	-3	0	94	106
5	-3	0	58	79
6	-3	0	364	334
7	-3	0	127	124
8	-3	0	36	46
9	-3	0	49	62
1	-2	0	132	120
2	-2	0	411	389
3	-2	0	722	782
4	-2	0	256	206
5	-2	0	112	117
6	-2	0	374	370
7	-2	0	144	137
8	-2	0	55	57
9	-2	0	46	39
10	-2	0	159	163
11	-2	0	80	89
12	-2	0	53	55
14	-2	0	46	45
2	-1	0	269	257
3	-1	0	825	917
4	-1	0	340	296
6	-1	0	108	107
7	-1	0	33	65
8	-1	0	46	43
11	-1	0	60	67
2	0	0	56	53
3	0	0	1006	1172
4	0	0	361	398
5	0	0	96	83
6	0	0	225	207
7	0	0	192	201
8	0	0	81	75
9	0	0	49	39
10	0	0	219	223
11	0	0	216	233
13	0	0	73	76
2	-14	1	16	26
6	-14	1	24	22
0	-13	1	10	30
4	-13	1	55	44
5	-13	1	26	19
1	-12	1	27	60
3	-12	1	22	20
4	-12	1	26	32
-4	-11	1	25	29
-3	-11	1	22	25
-2	-11	1	39	35
0	-11	1	10	56
1	-11	1	39	74
2	-11	1	18	9
5	-11	1	38	44
8	-11	1	29	45
-7	-10	1	38	34
-6	-10	1	83	66
-2	-10	1	34	60
-1	-10	1	19	49
0	-10	1	24	47
1	-10	1	19	32
2	-10	1	43	51
4	-10	1	26	25
5	-10	1	52	80
7	-10	1	35	45
11	-10	1	39	52
-7	-9	1	65	48
-6	-9	1	28	27
-5	-9	1	43	41
-4	-9	1	23	22
-2	-9	1	66	125
-1	-9	1	16	30
1	-9	1	17	26
2	-9	1	64	61
4	-9	1	51	51
5	-9	1	67	50
6	-9	1	34	28
7	-9	1	39	25
-11	-8	1	40	47
-9	-8	1	58	45
-7	-8	1	150	134
-6	-8	1	151	140
-5	-8	1	62	73
-4	-8	1	58	43
-2	-8	1	25	40

-1	-8	1	33	72
0	-8	1	40	79
1	-8	1	39	58
2	-8	1	82	84
4	-8	1	116	105
5	-8	1	71	84
6	-8	1	66	63
8	-8	1	43	48
9	-8	1	85	59
-15	-7	1	51	71
-11	-7	1	55	45
-9	-7	1	118	92
-8	-7	1	78	74
-7	-7	1	42	39
-6	-7	1	80	69
-4	-7	1	98	95
-3	-7	1	32	52
-2	-7	1	34	44
-1	-7	1	15	42
0	-7	1	84	148
1	-7	1	55	83
2	-7	1	102	113
3	-7	1	73	67
4	-7	1	67	79
5	-7	1	141	116
6	-7	1	34	37
7	-7	1	50	52
8	-7	1	53	55
-11	-6	1	77	72
-8	-6	1	46	45
-7	-6	1	111	110
-4	-6	1	50	71
-3	-6	1	158	166
-2	-6	1	126	176
-1	-6	1	113	201
0	-6	1	15	32
1	-6	1	101	122
2	-6	1	227	208
3	-6	1	48	58
4	-6	1	83	69
5	-6	1	45	51
6	-6	1	65	73
7	-6	1	53	61
9	-6	1	183	154
12	-6	1	52	49
-12	-5	1	139	138
-9	-5	1	390	326
-8	-5	1	51	46
-6	-5	1	105	85
-5	-5	1	51	59
-4	-5	1	111	119
-3	-5	1	71	65
-2	-5	1	43	77
-1	-5	1	131	205
0	-5	1	30	29
1	-5	1	75	104
2	-5	1	316	317
3	-5	1	141	137
4	-5	1	38	31
5	-5	1	98	84
6	-5	1	79	78
7	-5	1	58	67
8	-5	1	102	88
9	-5	1	100	89
11	-5	1	62	63
-11	-4	1	80	68
-10	-4	1	52	30
-9	-4	1	296	286
-8	-4	1	184	184
-7	-4	1	196	194
-6	-4	1	316	286
-5	-4	1	170	173
-4	-4	1	69	74
-3	-4	1	39	75
-2	-4	1	202	214
-1	-4	1	157	221
0	-4	1	132	211
1	-4	1	125	131
2	-4	1	69	77
3	-4	1	206	145
4	-4	1	126	133
6	-4	1	127	128
7	-4	1	116	117
8	-4	1	76	60
10	-4	1	71	74
11	-4	1	83	80
12	-4	1	73	64
13	-4	1	43	44
-12	-3	1	60	55
-11	-3	1	49	50
-10	-3	1	70	62
-9	-3	1	286	269
-8	-3	1	31	34
-7	-3	1	280	273
-6	-3	1	130	147
-5	-3	1	665	663
-4	-3	1	29	21
-3	-3	1	130	143
-2	-3	1	233	223
-1	-3	1	287	314
0	-3	1	86	96
1	-3	1	779	784
2	-3	1	720	719
3	-3	1	137	154
4	-3	1	212	195
5	-3	1	162	163
6	-3	1	54	61
7	-3	1	60	67
8	-3	1	98	85
9	-3	1	131	134
11	-3	1	40	42
-10	-2	1	53	82
-9	-2	1	141	123
-8	-2	1	63	51
-7	-2	1	272	265
-6	-2	1	199	179
-5	-2	1	390	384
-4	-2	1	542	522
-3	-2	1	638	649
-2	-2	1	445	474
-1	-2	1	254	267
0	-2	1	350	365
1	-2	1	741	758
2	-2	1	403	411
3	-2	1	341	329
4	-2	1	266	281
5	-2	1	30	26
6	-2	1	236	274
10	-2	1	140	151
12	-2	1	39	53
-12	-1	1	57	71
-11	-1	1	70	61
-10	-1	1	47	41
-9	-1	1	69	71
-7	-1	1	75	87
-6	-1	1	71	67
-5	-1	1	117	121
-4	-1	1	19	26
-3	-1	1	778	828
1	-1	1	399	404
2	-1	1	550	592
3	-1	1	148	143
4	-1	1	55	55
5	-1	1	421	371
6	-1	1	158	139
7	-1	1	252	249
8	-1	1	147	135
9	-1	1	253	252
13	-1	1	57	55
2	-14	2	19	26
-6	-13	2	25	21
0	-13	2	14	31
-3	-12	2	15	24
4	-12	2	32	37
5	-12	2	60	45
-10	-11	2	42	37
-5	-11	2	26	32
-3	-11	2	27	38
0	-11	2	27	52
1	-11	2	19	31
2	-11	2	39	42
3	-11	2	20	29

4 -11	2	54	53
-7 -10	2	64	58
-3 -10	2	48	72
-2 -10	2	19	43
-1 -10	2	37	68
0 -10	2	37	53
2 -10	2	25	27
3 -10	2	100	108
5 -10	2	53	41
6 -10	2	34	34
-8 -9	2	60	67
-7 -9	2	43	42
-6 -9	2	72	70
-5 -9	2	49	64
-4 -9	2	78	100
-3 -9	2	20	30
-2 -9	2	42	73
-1 -9	2	41	71
0 -9	2	16	17
2 -9	2	75	87
4 -9	2	70	79
9 -9	2	51	49
10 -9	2	67	50
-9 -8	2	85	72
-7 -8	2	44	46
-5 -8	2	77	70
-4 -8	2	106	126
-3 -8	2	48	52
-1 -8	2	34	49
0 -8	2	85	123
2 -8	2	53	48
3 -8	2	136	118
4 -8	2	64	72
5 -8	2	33	25
6 -8	2	58	54
7 -8	2	104	101
-14 -7	2	40	55
-12 -7	2	76	75
-8 -7	2	189	204
-7 -7	2	55	26
-6 -7	2	62	73
-5 -7	2	28	30
-4 -7	2	115	139
-3 -7	2	22	29
-2 -7	2	70	92
0 -7	2	35	71
1 -7	2	80	69
2 -7	2	44	50
3 -7	2	23	22
4 -7	2	80	92
5 -7	2	49	56
6 -7	2	222	209
7 -7	2	38	52
8 -7	2	62	71
13 -7	2	43	54
-14 -6	2	67	74
-12 -6	2	91	96
-11 -6	2	186	160
-10 -6	2	49	44
-9 -6	2	126	125
-8 -6	2	37	26
-7 -6	2	173	172
-6 -6	2	200	163
-5 -6	2	22	40
-4 -6	2	25	54
-3 -6	2	163	186
0 -6	2	213	231
1 -6	2	164	159
2 -6	2	66	67
3 -6	2	197	177
4 -6	2	177	158
5 -6	2	37	19
7 -6	2	83	102
8 -6	2	73	56
9 -6	2	95	95
10 -6	2	64	52
-12 -5	2	102	109
-10 -5	2	82	77
-9 -5	2	206	194
-8 -5	2	34	43
-7 -5	2	69	59
-6 -5	2	113	127
-5 -5	2	323	372
-4 -5	2	228	234
-3 -5	2	103	124
-2 -5	2	91	89
0 -5	2	68	127
1 -5	2	194	196
2 -5	2	256	234
3 -5	2	77	76
4 -5	2	88	79
5 -5	2	106	92
6 -5	2	111	125
7 -5	2	47	53
9 -5	2	122	112
-14 -4	2	67	67
-11 -4	2	172	155
-9 -4	2	52	62
-8 -4	2	58	61
-7 -4	2	195	174
-6 -4	2	62	65
-5 -4	2	116	126
-4 -4	2	258	253
-3 -4	2	202	186
-2 -4	2	36	57
-1 -4	2	140	158
0 -4	2	343	376
1 -4	2	171	184
2 -4	2	47	69
3 -4	2	123	134
4 -4	2	201	171
5 -4	2	157	140
7 -4	2	184	186
8 -4	2	87	89
9 -4	2	58	60
10 -4	2	65	62
13 -4	2	45	51
-11 -3	2	119	101
-10 -3	2	193	176
-9 -3	2	141	111
-8 -3	2	194	212
-7 -3	2	96	84
-6 -3	2	221	196
-5 -3	2	183	177
-4 -3	2	168	162
-3 -3	2	140	131
-2 -3	2	106	118
-1 -3	2	224	245
0 -3	2	329	301
1 -3	2	258	261
2 -3	2	305	290
3 -3	2	305	284
4 -3	2	130	173
5 -3	2	51	58
6 -3	2	43	59
7 -3	2	80	82
8 -3	2	97	102
9 -3	2	107	114
11 -3	2	49	55
12 -3	2	77	91
-12 -2	2	94	110
-10 -2	2	38	36
-9 -2	2	71	63
-8 -2	2	63	65
-7 -2	2	314	311
-6 -2	2	171	176
-5 -2	2	542	506
-4 -2	2	304	285
-3 -2	2	110	107
-2 -2	2	233	196
-1 -2	2	318	322
0 -2	2	571	585
1 -2	2	570	605
3 -2	2	74	79
4 -2	2	22	27
5 -2	2	39	36
6 -2	2	61	78
7 -2	2	150	159
9 -2	2	175	178
11 -2	2	124	132
13 -2	2	54	59
-11 -1	2	112	104

-10	-1	2	134	128
-9	-1	2	121	119
-7	-1	2	118	110
-6	-1	2	300	294
-5	-1	2	62	55
-4	-1	2	315	323
-3	-1	2	305	305
-2	-1	2	351	346
-1	-1	2	100	99
0	-1	2	499	520
1	-1	2	395	388
2	-1	2	490	453
3	-1	2	176	148
4	-1	2	62	55
5	-1	2	209	215
6	-1	2	54	72
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-12	-6	11	60	70	-6	-1	11	296	303	-4	-2	12	103	109
-8	-6	11	61	62	-5	-1	11	127	131	-1	-2	12	59	57
-7	-6	11	39	43	-4	-1	11	155	164	0	-2	12	118	118
-3	-6	11	91	75	-3	-1	11	102	109	1	-2	12	38	43
4	-6	11	51	64	-2	-1	11	67	64	2	-2	12	86	85
-10	-5	11	49	66	-1	-1	11	46	50	-12	-1	12	43	57
-8	-5	11	39	48	0	-1	11	95	97	-6	-1	12	38	39
-5	-5	11	65	80	1	-1	11	154	153	-4	-1	12	85	87
-4	-5	11	57	55	3	-1	11	40	44	-1	-1	12	51	61
-3	-5	11	89	82	4	-1	11	47	54	2	-1	12	80	91
-2	-5	11	60	53	5	-1	11	51	53	-14	0	12	63	78
1	-5	11	118	118	-5	-11	12	83	72	-12	0	12	68	79
4	-5	11	46	71	-5	-9	12	55	45	-8	0	12	102	106
7	-5	11	43	56	-4	-9	12	66	45	-7	0	12	71	67
-12	-4	11	50	58	-7	-8	12	45	35	-6	0	12	101	106
-9	-4	11	47	53	-4	-8	12	41	57	-5	0	12	138	160
-5	-4	11	61	59	-2	-8	12	59	46	-4	0	12	91	79
-2	-4	11	96	92	2	-8	12	54	56	-3	0	12	135	132
-1	-4	11	40	54	-7	-7	12	76	75	-2	0	12	62	62
2	-4	11	64	66	-6	-7	12	87	93	-1	0	12	254	260
6	-4	11	42	59	-2	-7	12	63	47	0	0	12	92	105
-12	-3	11	63	75	-1	-7	12	87	93	1	0	12	68	78
-11	-3	11	53	64	0	-7	12	61	76	4	0	12	43	51
-10	-3	11	55	66	1	-7	12	55	54	6	0	12	51	68
-6	-3	11	119	127	3	-7	12	43	53	-3	-9	13	86	76
-5	-3	11	100	96	-9	-6	12	68	65	-12	-8	13	50	58
-4	-3	11	42	39	-4	-6	12	105	94	-10	-8	13	49	56
-3	-3	11	127	119	-3	-6	12	38	39	-7	-8	13	62	59
-2	-3	11	85	90	4	-6	12	39	42	-5	-8	13	74	62
-1	-3	11	74	70	6	-6	12	64	64	-10	-7	13	41	54
1	-3	11	51	46	-14	-5	12	53	69	1	-7	13	55	65
2	-3	11	117	125	-7	-5	12	45	61	-7	-6	13	60	64
3	-3	11	51	54	-6	-5	12	62	55	-4	-6	13	42	43
6	-3	11	55	63	-5	-5	12	89	86	-3	-6	13	89	95
-14	-2	11	49	66	-3	-5	12	43	40	-10	-5	13	71	86
-13	-2	11	51	55	4	-5	12	82	84	-8	-5	13	50	62
-12	-2	11	76	93	-6	-4	12	49	51	-7	-5	13	40	34
-9	-2	11	132	139	-4	-4	12	76	68	-6	-5	13	131	140
-7	-2	11	41	36	-3	-4	12	41	41	-3	-5	13	95	105
-6	-2	11	40	33	0	-4	12	43	49	1	-5	13	76	96
-5	-2	11	116	116	6	-4	12	46	57	3	-5	13	73	88
-4	-2	11	69	74	-12	-3	12	57	62	-9	-4	13	45	71
-3	-2	11	143	137	-8	-3	12	44	40	-6	-4	13	84	83
-2	-2	11	105	106	-7	-3	12	75	80	-4	-4	13	56	57

-3	-4	13	84	94
0	-4	13	62	60
1	-4	13	49	59
3	-4	13	56	72
-13	-3	13	58	66
-12	-3	13	53	71
-8	-3	13	89	102
-5	-3	13	50	46
-4	-3	13	56	69
-3	-3	13	66	78
0	-3	13	109	120
1	-3	13	52	58
-4	-2	13	89	97
0	-2	13	54	58
-9	-1	13	44	55
-5	-1	13	76	82
-3	-1	13	44	57
-2	-1	13	113	135
-1	-1	13	38	46
0	-1	13	113	125
2	-1	13	65	79
3	-1	13	42	48
4	-1	13	56	61
-5	-8	14	46	42
-10	-7	14	73	85
-7	-7	14	55	63
-2	-7	14	96	105
-10	-6	14	70	81
-9	-6	14	48	58
-2	-6	14	48	51
1	-6	14	113	129
-11	-5	14	49	54
-5	-5	14	60	67
-9	-4	14	77	96
-6	-4	14	40	35
-2	-4	14	52	73
0	-4	14	61	78
1	-4	14	114	138
-5	-3	14	134	151
-4	-3	14	49	65
-3	-3	14	45	49
-2	-3	14	82	98
-1	-3	14	44	57
-12	-2	14	43	64
-9	-2	14	61	80
0	-2	14	47	60
-12	-1	14	42	53
-5	-1	14	54	58
-12	0	14	53	60
-5	0	14	47	44
2	0	14	43	46
-3	-8	15	69	71
-2	-7	15	65	74
-4	-6	15	73	75
-3	-6	15	98	115
-2	-5	15	46	54
-1	-5	15	115	138
0	-5	15	59	69
-3	-4	15	55	63
-1	-4	15	42	64
-3	-3	15	45	57
0	-3	15	51	61
1	-3	15	42	59
0	-2	15	61	77
-8	-1	15	66	91
-5	-1	15	40	60
-1	-1	15	43	58

Table 2. Anistropic thermal parameters ($/\text{Å}^2 \times 10^3$).

	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}
C(1)	36(6)	50(8)	22(5)	-9(5)	4(4)	19(6)
O(11)	42(5)	103(9)	49(6)	-30(6)	16(4)	-35(6)
N(11)	33(5)	70(8)	27(5)	25(5)	16(4)	20(5)
N(12)	32(6)	84(10)	44(6)	-16(7)	13(5)	-2(6)
C(11)	32(6)	53(8)	22(5)	15(5)	12(4)	11(6)
C(12)	51(8)	62(9)	30(6)	5(6)	19(6)	7(7)
C(13)	23(5)	56(9)	26(5)	-3(6)	-2(4)	-13(6)
C(14)	61(10)	104(14)	40(7)	5(9)	37(7)	-13(10)
C(15)	66(9)	63(10)	23(6)	-11(6)	17(6)	-13(8)
C(16)	57(9)	139(18)	25(6)	3(8)	23(6)	-23(11)
O(21)	44(5)	66(6)	37(5)	12(5)	6(4)	23(5)
N(21)	35(6)	67(8)	24(5)	2(5)	9(4)	-19(5)
N(22)	29(5)	57(7)	14(4)	3(4)	5(3)	-6(4)
C(21)	41(7)	48(7)	18(5)	9(5)	7(4)	22(6)
C(22)	29(7)	86(12)	39(7)	-27(8)	5(5)	-11(7)
C(23)	36(6)	38(7)	15(5)	-5(4)	11(4)	-5(5)
C(24)	52(8)	53(9)	33(6)	5(6)	20(6)	18(7)
C(25)	65(9)	102(12)	18(6)	30(7)	16(6)	39(9)
C(26)	60(10)	140(21)	45(9)	-47(11)	5(7)	-25(12)
C(3)	32(6)	67(9)	19(5)	11(5)	5(4)	-29(6)
O(31)	35(5)	139(12)	56(6)	-11(7)	9(5)	-42(7)
N(31)	32(5)	88(9)	37(5)	-46(6)	20(4)	-31(6)
N(32)	40(6)	63(7)	20(4)	-17(5)	14(4)	-23(5)
C(31)	23(5)	70(9)	18(5)	-13(5)	9(4)	-26(6)
C(32)	28(6)	94(12)	33(6)	-19(7)	11(5)	-35(7)
C(33)	40(7)	63(8)	20(5)	-1(6)	17(5)	13(6)
C(34)	42(8)	140(17)	36(7)	-43(9)	20(6)	9(9)
C(35)	63(11)	168(22)	57(9)	-72(12)	20(8)	-34(13)
C(36)	23(6)	71(9)	40(6)	-12(7)	20(5)	5(6)
O(41)	65(7)	102(10)	41(5)	28(6)	8(5)	3(7)
N(41)	37(5)	55(7)	22(4)	-1(4)	7(4)	19(5)
N(42)	44(6)	87(10)	22(5)	5(6)	13(4)	24(6)
C(41)	27(6)	62(9)	15(4)	-4(5)	0(4)	-18(6)
C(42)	29(6)	51(8)	18(4)	28(5)	4(4)	-1(5)
C(43)	26(6)	78(10)	26(5)	5(6)	7(5)	-6(6)
C(44)	37(7)	88(11)	47(8)	-40(8)	4(6)	-16(7)
C(45)	51(8)	121(15)	17(5)	-29(7)	14(5)	-37(9)
C(46)	38(6)	55(8)	26(5)	10(5)	15(5)	30(6)

The anisotropic temperature factor takes the form

$$\exp\left[-2\pi^2(h^2\underline{a}^{*2}U_{11} + k^2\underline{b}^{*2}U_{22} + l^2\underline{c}^{*2}U_{33} + 2kl\underline{b}^*\underline{c}^*U_{23} + 2lh\underline{c}^*\underline{a}^*U_{31} + 2hk\underline{a}^*\underline{b}^*U_{12}\right]$$

The common isotropic temperature factor for the methyl hydrogen atoms refined to 140(13) x 10^{-3} Å^2; that for the methylene hydrogen atoms to 29(40) x 10^{-3} Å^2.

IV. 4-Phenyl-3-phenylamino-1,2,4-thiadiazoline-5-one.

(i) Structure Factors.

(ii) Thermal Parameters.

OBSERVED AND CALCULATED STRUCTURE FACTORS FOR DOST'S KETO COMPOUND

H	K	L	10FO	10FC
2	0	0	586	612
4	0	0	143	129
6	0	0	74	53
1	1	0	156	144
2	1	0	373	355
3	1	0	367	356
4	1	0	122	137
6	1	0	328	313
7	1	0	188	202
8	1	0	152	147
9	1	0	132	122
10	1	0	167	171
12	1	0	45	58
1	2	0	127	131
2	2	0	77	93
3	2	0	503	482
5	2	0	91	84
7	2	0	91	91
11	2	0	99	89
12	2	0	77	92
14	2	0	48	45
1	3	0	422	499
2	3	0	345	332
3	3	0	387	401
4	3	0	32	52
6	3	0	127	133
7	3	0	144	154
9	3	0	61	54
13	3	0	109	124
15	3	0	65	78
0	4	0	20	83
1	4	0	216	262
2	4	0	24	48
3	4	0	236	237
4	4	0	410	397
5	4	0	108	112
6	4	0	189	178
8	4	0	116	123
9	4	0	71	78
10	4	0	90	77
11	4	0	57	44
1	5	0	243	340
2	5	0	213	214
4	5	0	371	339
6	5	0	91	105
7	5	0	79	73
8	5	0	70	75
10	5	0	227	205
11	5	0	125	129
0	6	0	26	95
1	6	0	29	33
2	6	0	29	19
3	6	0	185	178
5	6	0	266	272
6	6	0	76	80
7	6	0	106	101
8	6	0	65	79
9	6	0	120	146
10	6	0	93	83
13	6	0	82	80
1	7	0	135	204
3	7	0	189	186
4	7	0	174	161
6	7	0	146	143
7	7	0	77	93
8	7	0	41	41
11	7	0	70	72
12	7	0	101	98
1	8	0	18	41
2	8	0	87	98
4	8	0	161	169
5	8	0	91	100
6	8	0	42	40
8	8	0	78	90
9	8	0	47	63
14	8	0	76	65
2	9	0	59	77
3	9	0	100	99
4	9	0	142	145
5	9	0	83	83
6	9	0	59	50
10	9	0	48	37
12	9	0	67	57
0	10	0	21	120
1	10	0	25	44
3	10	0	55	81
4	10	0	45	57
6	10	0	35	33
7	10	0	113	115
9	10	0	91	87
11	10	0	79	61
1	11	0	54	124
2	11	0	31	54
3	11	0	30	51
4	11	0	58	41
5	11	0	114	114
0	12	0	145	76
1	12	0	40	90
2	12	0	41	43
3	12	0	22	29
5	12	0	61	63
7	12	0	34	28
8	12	0	56	55
1	13	0	21	38
4	13	0	59	64
10	13	0	111	70
1	14	0	31	54
5	14	0	68	65
1	0	1	386	382
2	0	1	1106	1182
3	0	1	229	212
4	0	1	371	373
5	0	1	335	319
6	0	1	445	423
8	0	1	167	187
13	0	1	48	67
0	1	1	371	363
1	1	1	334	314
2	1	1	792	830
3	1	1	439	426
4	1	1	279	300
5	1	1	265	256
6	1	1	429	396
7	1	1	139	153
8	1	1	85	94
10	1	1	73	66
11	1	1	111	104
12	1	1	113	118
13	1	1	81	98
0	2	1	194	189
1	2	1	718	730
2	2	1	691	708
3	2	1	745	749
4	2	1	464	467
5	2	1	127	132
6	2	1	179	169
7	2	1	167	181
11	2	1	135	139
12	2	1	140	137
15	2	1	59	75
0	3	1	184	225
1	3	1	744	786
2	3	1	398	411
3	3	1	95	94
4	3	1	184	173
5	3	1	359	350
6	3	1	130	116
7	3	1	161	160
8	3	1	94	113
9	3	1	58	45
10	3	1	47	39
11	3	1	161	164
13	3	1	88	93
14	3	1	59	70
0	4	1	69	80
1	4	1	277	290
2	4	1	151	148
3	4	1	281	258
4	4	1	82	67
5	4	1	287	276
6	4	1	169	155
7	4	1	89	89

0	5	1	14	23
1	5	1	96	120
2	5	1	246	254
3	5	1	245	207
4	5	1	158	155
5	5	1	70	82
6	5	1	170	150
7	5	1	142	139
8	5	1	162	186
9	5	1	158	132
10	5	1	137	119
11	5	1	54	53
0	6	1	60	95
1	6	1	363	455
2	6	1	136	132
3	6	1	163	170
4	6	1	133	151
5	6	1	74	48
6	6	1	190	184
7	6	1	60	60
8	6	1	56	55
10	6	1	57	48
11	6	1	84	76
12	6	1	56	61
0	7	1	148	225
1	7	1	156	176
2	7	1	93	123
3	7	1	56	49
4	7	1	87	94
5	7	1	127	115
7	7	1	115	112
8	7	1	58	76
9	7	1	102	117
11	7	1	121	110
12	7	1	57	54
13	7	1	50	46
1	8	1	28	40
2	8	1	128	132
3	8	1	51	57
4	8	1	102	101
5	8	1	90	94
6	8	1	241	222
8	8	1	53	47
9	8	1	34	45
10	8	1	75	64
10	9	1	58	96
2	9	1	139	183
4	9	1	58	66
5	9	1	61	57
6	9	1	97	81
7	9	1	83	78
9	9	1	82	92
0	10	1	56	87
1	10	1	72	104
2	10	1	47	69
3	10	1	75	84
4	10	1	96	105
5	10	1	40	28
6	10	1	44	55
7	10	1	63	63
0	11	1	62	106
1	11	1	43	86
2	11	1	54	60
3	11	1	38	42
7	11	1	104	95
8	11	1	43	44
11	11	1	75	57
1	12	1	16	39
2	12	1	27	38
3	12	1	53	61
4	12	1	63	65
5	12	1	37	30
6	12	1	64	69
8	12	1	60	48
10	12	1	86	57
1	13	1	27	39
2	13	1	56	78
3	13	1	28	26
5	13	1	55	53
8	13	1	42	38
1	14	1	28	37
0	0	2	165	159
1	0	2	566	580
2	0	2	525	522
3	0	2	100	95
4	0	2	557	556
5	0	2	236	240
6	0	2	120	108
7	0	2	101	95
10	0	2	78	84
11	0	2	55	53
0	1	2	491	484
1	1	2	318	304
2	1	2	215	205
3	1	2	497	484
4	1	2	342	312
5	1	2	168	167
6	1	2	240	224
7	1	2	157	154
10	1	2	87	80
12	1	2	48	54
0	2	2	172	164
1	2	2	1014	1056
2	2	2	694	708
3	2	2	293	284
4	2	2	322	312
5	2	2	355	375
6	2	2	360	334
7	2	2	239	251
11	2	2	73	75
12	2	2	45	41
13	2	2	64	79
14	2	2	84	87
0	3	2	27	33
1	3	2	350	337
2	3	2	193	168
3	3	2	119	119
4	3	2	223	211
5	3	2	408	395
6	3	2	360	313
7	3	2	104	111
8	3	2	121	131
9	3	2	89	77
10	3	2	74	78
13	3	2	76	83
0	4	2	438	434
1	4	2	223	198
2	4	2	39	41
3	4	2	88	85
4	4	2	235	220
5	4	2	53	55
6	4	2	189	169
7	4	2	36	43
8	4	2	40	46
9	4	2	68	58
11	4	2	91	85
12	4	2	60	60
13	4	2	74	78
14	4	2	65	72
0	5	2	104	87
1	5	2	131	128
2	5	2	205	201
3	5	2	412	394
5	5	2	166	160
6	5	2	85	71
7	5	2	38	40
8	5	2	236	266
9	5	2	152	133
10	5	2	127	122
11	5	2	48	47
12	5	2	91	84
0	6	2	156	154
1	6	2	111	101
2	6	2	91	88
3	6	2	420	408
4	6	2	106	99
5	6	2	93	89
6	6	2	75	76
7	6	2	227	233
8	6	2	138	142
9	6	2	141	120
10	6	2	125	99

0	7	2	183	204
1	7	2	188	212
2	7	2	138	147
3	7	2	32	34
4	7	2	188	192
5	7	2	121	126
6	7	2	65	63
8	7	2	56	52
10	7	2	68	61
11	7	2	86	79
12	7	2	51	55
13	7	2	61	61
0	8	2	69	84
1	8	2	85	88
2	8	2	210	217
3	8	2	98	110
4	8	2	93	97
5	8	2	36	44
6	8	2	80	67
7	8	2	94	95
8	8	2	78	92
9	8	2	42	35
12	8	2	59	57
13	8	2	47	37
0	9	2	72	105
1	9	2	60	76
2	9	2	73	73
3	9	2	49	48
4	9	2	159	153
5	9	2	31	34
6	9	2	96	81
7	9	2	78	71
8	9	2	72	70
9	9	2	69	50
11	9	2	92	74
0	10	2	66	83
1	10	2	24	33
2	10	2	117	141
3	10	2	48	53
4	10	2	57	54
5	10	2	104	105
6	10	2	31	25
7	10	2	34	37
9	10	2	53	48
0	11	2	64	74
1	11	2	31	43
2	11	2	20	21
3	11	2	49	34
4	11	2	49	53
5	11	2	103	103
8	11	2	75	76
9	11	2	46	41
1	12	2	42	55
2	12	2	52	61
3	12	2	37	37
1	13	2	27	28
2	13	2	30	36
4	13	2	48	51
5	13	2	43	41
7	13	2	50	44
6	14	2	32	32
7	14	2	69	59
1	0	3	58	50
2	0	3	486	442
3	0	3	96	78
4	0	3	41	37
5	0	3	389	378
6	0	3	373	366
7	0	3	168	189
15	0	3	72	88
0	1	3	242	237
1	1	3	173	162
2	1	3	453	409
3	1	3	203	175
4	1	3	386	385
5	1	3	408	384
6	1	3	260	251
7	1	3	132	115
9	1	3	138	133
11	1	3	134	132
12	1	3	94	97
14	1	3	56	67
1	2	3	246	252
2	2	3	488	427
3	2	3	360	328
4	2	3	464	465
5	2	3	101	91
9	2	3	69	67
11	2	3	75	73
14	2	3	70	92
15	2	3	53	67
0	3	3	73	43
1	3	3	181	184
2	3	3	127	87
3	3	3	348	317
4	3	3	304	288
5	3	3	312	294
6	3	3	231	232
7	3	3	189	205
8	3	3	72	86
10	3	3	102	102
11	3	3	85	85
12	3	3	52	64
13	3	3	46	50
14	3	3	52	74
0	4	3	65	55
2	4	3	371	321
3	4	3	215	188
4	4	3	215	221
5	4	3	241	222
6	4	3	274	260
8	4	3	101	112
12	4	3	78	82
13	4	3	76	68
14	4	3	58	62
0	5	3	208	190
1	5	3	382	361
2	5	3	210	180
3	5	3	80	71
4	5	3	202	194
5	5	3	196	166
6	5	3	127	126
7	5	3	135	146
8	5	3	104	116
9	5	3	143	124
10	5	3	113	106
11	5	3	48	47
0	6	3	339	330
1	6	3	169	167
2	6	3	77	75
3	6	3	174	157
4	6	3	111	101
5	6	3	134	112
6	6	3	112	107
7	6	3	85	106
8	6	3	125	133
9	6	3	234	200
0	7	3	130	133
1	7	3	111	111
2	7	3	198	196
4	7	3	70	60
5	7	3	197	177
6	7	3	184	176
8	7	3	57	57
10	7	3	62	55
13	7	3	64	74
0	8	3	57	50
1	8	3	92	106
2	8	3	79	76
3	8	3	47	50
4	8	3	55	52
5	8	3	132	135
6	8	3	216	202
7	8	3	51	48
8	8	3	96	94
9	8	3	117	88
11	8	3	65	53
1	9	3	76	76
2	9	3	165	185
3	9	3	158	169
4	9	3	103	104
5	9	3	59	44

6	9	3	93	94
7	9	3	46	37
9	9	3	75	69
10	9	3	86	58
0	10	3	144	145
1	10	3	53	66
2	10	3	28	39
3	10	3	81	75
4	10	3	132	133
5	10	3	73	45
1	11	3	55	51
3	11	3	34	31
4	11	3	30	28
6	11	3	74	60
7	11	3	59	54
11	11	3	63	55
3	12	3	26	23
5	12	3	48	36
6	12	3	51	41
7	12	3	41	32
1	13	3	33	45
2	13	3	29	40
3	13	3	35	26
5	13	3	41	45
7	13	3	30	24
4	14	3	41	34
0	0	4	592	593
1	0	4	172	178
2	0	4	102	113
3	0	4	501	445
4	0	4	153	136
6	0	4	29	44
9	0	4	51	48
10	0	4	45	54
11	0	4	69	75
12	0	4	50	43
0	1	4	295	282
1	1	4	319	304
2	1	4	169	176
3	1	4	79	65
4	1	4	324	284
5	1	4	44	49
6	1	4	194	200
7	1	4	96	101
8	1	4	107	111
11	1	4	51	67
13	1	4	49	50
0	2	4	251	226
1	2	4	174	161
2	2	4	181	175
3	2	4	272	239
4	2	4	281	270
5	2	4	93	80
7	2	4	32	36
0	3	4	52	41
1	3	4	283	262
2	3	4	302	290
3	3	4	225	200
4	3	4	280	254
5	3	4	153	162
6	3	4	85	95
7	3	4	228	253
9	3	4	127	123
10	3	4	111	120
11	3	4	87	88
13	3	4	51	56
0	4	4	262	257
1	4	4	217	210
2	4	4	133	142
3	4	4	348	310
4	4	4	171	169
5	4	4	288	286
7	4	4	43	63
8	4	4	178	144
10	4	4	53	54
12	4	4	70	78
0	5	4	123	139
1	5	4	114	99
2	5	4	193	188
3	5	4	119	92
5	5	4	37	41
6	5	4	73	67
7	5	4	301	325
8	5	4	124	104
9	5	4	72	60
10	5	4	119	122
11	5	4	75	82
13	5	4	61	72
0	6	4	100	99
1	6	4	74	61
2	6	4	54	61
3	6	4	91	71
4	6	4	172	150
5	6	4	247	234
6	6	4	106	99
7	6	4	66	86
8	6	4	167	134
13	6	4	54	64
14	6	4	61	64
0	7	4	150	167
1	7	4	158	170
2	7	4	108	112
3	7	4	138	113
4	7	4	229	201
5	7	4	172	166
6	7	4	218	215
7	7	4	131	125
11	7	4	60	52
0	8	4	94	98
1	8	4	75	76
2	8	4	94	99
3	8	4	334	283
4	8	4	230	196
5	8	4	69	58
6	8	4	45	33
10	8	4	64	57
12	8	4	45	46
0	9	4	54	48
1	9	4	82	73
2	9	4	114	116
3	9	4	53	43
4	9	4	143	124
5	9	4	122	109
6	9	4	62	60
7	9	4	48	41
8	9	4	66	54
9	9	4	76	52
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1	10	4	35	42
2	10	4	38	43
3	10	4	75	91
4	10	4	76	61
6	10	4	101	99
7	10	4	38	34
8	10	4	76	57
9	10	4	46	37
0	11	4	71	70
2	11	4	73	85
3	11	4	29	35
5	11	4	59	56
6	11	4	57	56
7	11	4	37	27
0	12	4	47	55
1	12	4	50	53
3	12	4	40	41
5	12	4	42	28
10	12	4	46	35
3	14	4	29	31
6	14	4	38	29
1	0	5	192	192
2	0	5	192	180
3	0	5	148	154
6	0	5	131	158
11	0	5	106	126
15	0	5	56	73
0	1	5	206	196
1	1	5	285	265
2	1	5	158	155
3	1	5	183	177
4	1	5	168	179
5	1	5	106	112
7	1	5	102	93

10	1	5	71	86
11	1	5	60	60
13	1	5	53	69
0	2	5	96	132
1	2	5	107	107
2	2	5	474	435
3	2	5	191	196
4	2	5	168	152
7	2	5	104	107
8	2	5	56	57
10	2	5	61	75
11	2	5	60	56
0	3	5	54	43
1	3	5	332	322
2	3	5	158	152
3	3	5	231	216
4	3	5	316	319
5	3	5	142	138
6	3	5	125	141
7	3	5	57	57
8	3	5	158	151
9	3	5	51	51
10	3	5	44	42
12	3	5	71	76
14	3	5	48	55
0	4	5	54	55
1	4	5	92	87
2	4	5	90	91
3	4	5	101	90
6	4	5	45	57
10	4	5	99	106
11	4	5	65	75
14	4	5	54	63
1	5	5	141	135
2	5	5	99	103
3	5	5	153	156
4	5	5	148	153
5	5	5	116	122
7	5	5	89	66
8	5	5	193	181
10	5	5	99	87
12	5	5	66	75
14	5	5	54	52
0	6	5	163	156
1	6	5	101	78
2	6	5	214	184
3	6	5	215	210
5	6	5	210	216
6	6	5	96	110
8	6	5	69	60
10	6	5	59	54
11	6	5	47	46
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2	7	5	95	90
3	7	5	203	185
4	7	5	148	150
5	7	5	131	117
6	7	5	98	103
12	7	5	55	44
13	7	5	72	75
0	8	5	104	112
1	8	5	91	79
2	8	5	132	115
3	8	5	114	122
4	8	5	45	49
5	8	5	99	96
6	8	5	85	79
7	8	5	80	55
9	8	5	63	56
11	8	5	54	57
0	9	5	93	78
1	9	5	80	78
2	9	5	96	84
3	9	5	180	153
4	9	5	99	97
6	9	5	61	53
8	9	5	60	50
9	9	5	44	42
0	10	5	127	128
1	10	5	70	63
2	10	5	86	72
4	10	5	51	36
6	10	5	71	78
8	10	5	48	20
0	11	5	57	56
1	11	5	63	59
2	11	5	59	60
3	11	5	53	55
4	11	5	54	47
6	11	5	44	31
8	11	5	60	41
0	12	5	41	37
3	12	5	58	57
7	12	5	49	36
1	13	5	40	29
3	13	5	46	45
3	0	6	54	41
7	0	6	81	82
0	1	6	109	104
1	1	6	57	61
8	1	6	51	51
9	1	6	63	67
11	1	6	103	123
14	1	6	54	66
2	2	6	93	110
3	2	6	175	194
10	2	6	74	87
11	2	6	60	71
12	2	6	68	77
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1	3	6	134	149
2	3	6	210	227
3	3	6	108	112
4	3	6	158	178
5	3	6	255	220
6	3	6	141	121
7	3	6	83	77
8	3	6	65	65
9	3	6	96	93
11	3	6	62	76
4	4	6	111	125
5	4	6	115	106
6	4	6	97	90
9	4	6	52	39
0	5	6	32	37
1	5	6	232	235
2	5	6	194	202
3	5	6	109	130
5	5	6	227	179
6	5	6	107	106
7	5	6	122	103
8	5	6	74	74
9	5	6	157	150
10	5	6	81	83
11	5	6	59	66
0	6	6	36	37
1	6	6	55	57
2	6	6	132	146
3	6	6	122	132
4	6	6	69	73
5	6	6	61	33
7	6	6	62	52
8	6	6	111	103
10	6	6	149	146
0	7	6	185	173
1	7	6	71	85
2	7	6	158	147
3	7	6	61	64
5	7	6	59	51
6	7	6	71	60
7	7	6	118	100
8	7	6	69	55
11	7	6	72	79
12	7	6	69	72
0	8	6	125	116
1	8	6	113	107
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4	8	6	64	54
5	8	6	110	88
9	8	6	52	44
11	8	6	49	60
0	9	6	176	175

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3	9	6	64	54
4	9	6	71	73
5	9	6	72	84
6	9	6	62	47
7	9	6	56	37
9	9	6	62	47
2	10	6	105	102
3	10	6	40	47
5	10	6	42	47
6	10	6	101	73
0	11	6	97	81
1	11	6	35	31
3	11	6	35	26
4	11	6	56	64
2	12	6	42	48
3	12	6	49	45
6	14	6	34	33
2	0	7	96	89
3	0	7	60	54
5	0	7	91	90
0	1	7	44	30
4	1	7	159	143
7	1	7	91	91
10	1	7	111	122
13	1	7	53	77
8	2	7	80	86
10	2	7	60	65
12	2	7	57	68
13	2	7	73	74
0	3	7	172	163
1	3	7	148	129
2	3	7	142	115
3	3	7	86	73
4	3	7	97	89
5	3	7	105	88
6	3	7	122	125
7	3	7	91	96
8	3	7	94	91
9	3	7	61	56
11	3	7	62	63
5	4	7	201	197
6	4	7	69	71
9	4	7	70	82
11	4	7	69	78
0	5	7	184	149
1	5	7	174	153
3	5	7	166	155
4	5	7	84	63
5	5	7	121	122
7	5	7	69	69
8	5	7	129	125
9	5	7	46	44
10	5	7	79	84
0	6	7	148	111
1	6	7	176	152
2	6	7	146	126
3	6	7	137	111
4	6	7	72	64
7	6	7	73	63
8	6	7	84	79
12	6	7	53	55
0	7	7	49	33
1	7	7	132	96
2	7	7	91	56
3	7	7	109	96
5	7	7	125	111
6	7	7	75	65
8	7	7	46	42
9	7	7	59	52
10	7	7	54	62
0	8	7	48	52
2	8	7	50	33
3	8	7	46	26
4	8	7	60	50
7	8	7	69	73
9	8	7	82	74
11	8	7	53	54
12	8	7	61	60
1	9	7	73	69
2	9	7	84	55
3	9	7	69	63
6	9	7	59	58
7	9	7	49	47
9	9	7	55	49
0	10	7	89	75
1	10	7	53	53
2	10	7	91	97
0	11	7	49	50
2	11	7	73	66
6	11	7	69	49
0	12	7	63	74
4	12	7	45	33
5	12	7	77	59
1	0	8	72	67
9	0	8	50	38
13	0	8	87	118
1	1	8	159	139
5	1	8	58	50
7	1	8	84	93
8	1	8	62	62
9	1	8	75	81
1	2	8	67	64
2	2	8	95	101
6	2	8	60	69
7	2	8	58	63
8	2	8	145	165
10	2	8	56	61
11	2	8	64	83
1	3	8	47	52
2	3	8	145	143
3	3	8	185	186
4	3	8	94	104
6	3	8	105	111
7	3	8	93	88
8	3	8	56	66
11	3	8	63	54
0	4	8	47	46
1	4	8	189	170
3	4	8	96	104
6	4	8	63	66
9	4	8	81	85
1	5	8	101	97
2	5	8	55	48
3	5	8	44	42
4	5	8	106	118
5	5	8	122	115
7	5	8	102	99
10	5	8	83	95
1	6	8	129	127
2	6	8	52	37
4	6	8	60	60
6	6	8	86	78
7	6	8	49	38
8	6	8	68	66
9	6	8	60	64
2	7	8	87	66
3	7	8	70	70
4	7	8	91	91
5	7	8	70	74
9	7	8	67	61
0	8	8	84	56
3	8	8	63	54
5	8	8	73	79
6	8	8	53	51
1	9	8	72	48
7	9	8	50	29
2	10	8	51	36
6	10	8	72	64
7	10	8	80	79
0	11	8	55	43
2	11	8	63	44
1	12	8	62	54
2	12	8	69	54
5	12	8	51	51
1	0	9	71	60
4	0	9	48	39
6	0	9	57	68
7	0	9	130	147
10	0	9	61	78
3	1	9	111	111

5	1	9	51	55
6	1	9	55	66
8	1	9	62	66
9	1	9	100	118
0	2	9	68	79
1	2	9	176	183
3	2	9	83	97
4	2	9	80	92
8	2	9	61	76
9	2	9	57	64
1	3	9	96	112
4	3	9	86	104
6	3	9	73	93
7	3	9	70	95
9	3	9	66	75
0	4	9	106	107
3	4	9	70	59
4	4	9	73	87
5	4	9	67	76
6	4	9	66	82
7	4	9	86	96
10	4	9	68	75
1	5	9	81	77
2	5	9	86	90
3	5	9	77	72
4	5	9	58	55
9	5	9	59	64
1	6	9	68	58
6	6	9	55	59
7	6	9	58	56
8	6	9	59	65
4	7	9	86	94
7	8	9	79	74
0	9	9	74	69
3	9	9	56	48
4	9	9	86	77
2	10	9	57	38
5	10	9	68	63
1	11	9	69	56
2	11	9	54	33
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6	0	10	78	93
7	0	10	96	103
3	1	10	106	126
4	1	10	69	80
5	1	10	69	57
10	1	10	52	72
11	1	10	61	73
0	2	10	65	68
1	2	10	48	57
3	2	10	52	60
4	2	10	54	63
5	2	10	101	132
7	2	10	52	67
11	2	10	58	68
0	3	10	102	119
1	3	10	135	155
3	3	10	77	81
5	3	10	76	81
6	3	10	76	93
7	3	10	52	62
8	3	10	48	54
0	4	10	68	73
1	4	10	65	73
2	4	10	68	71
3	4	10	49	53
4	4	10	88	98
5	4	10	50	55
6	4	10	52	49
1	5	10	104	110
3	5	10	59	75
8	5	10	61	64
5	7	10	73	73
6	7	10	67	78
2	8	10	70	70
4	8	10	47	44
0	10	10	89	71
4	10	10	57	43
1	11	10	46	41
6	0	11	66	79
1	1	11	67	75
4	1	11	61	78
3	2	11	90	101
1	3	11	115	136
2	3	11	61	59
4	3	11	55	54
5	3	11	56	72
6	4	11	71	74
0	5	11	85	109
3	5	11	53	60
2	7	11	49	51
7	7	11	49	57
1	8	11	52	57
4	8	11	69	65
0	0	12	54	71
2	2	12	60	63
2	3	12	57	68
3	3	12	62	88
1	5	12	59	66
3	5	12	55	64
0	6	12	59	74
0	7	12	74	84
0	1	13	63	83

TABLE 2 Anisotropic thermal parameters ($/\AA^2 \times 10^3$)

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{31}	U_{12}
S(1)	48(2)	62(2)	42(2)	-2(2)	-13(2)	-10(2)
N(1)	38(6)	63(6)	22(5)	-1(4)	-7(4)	-4(5)
N(2)	27(5)	52(5)	23(5)	5(4)	-9(4)	0(4)
N(3)	26(5)	59(6)	30(5)	8(5)	-6(5)	-8(4)
O(1)	51(6)	63(6)	38(5)	-22(4)	-3(5)	-5(5)
C(1)	34(6)	37(5)	26(5)	-5(4)	-1(5)	9(5)
C(2)	30(7)	79(9)	42(7)	-29(7)	8(6)	-2(7)
C(3)	35(7)	47(6)	26(6)	0(5)	0(5)	9(5)
C(4)	37(8)	61(8)	51(9)	17(7)	-10(7)	0(6)
C(5)	46(9)	99(12)	41(9)	20(8)	-15(7)	0(8)
C(6)	81(11)	68(8)	28(6)	19(7)	-9(7)	22(9)
C(7)	58(9)	68(8)	41(7)	9(7)	7(7)	-15(8)
C(8)	47(8)	57(7)	30(7)	7(6)	0(6)	-3(6)
C(9)	33(6)	42(5)	23(5)	4(5)	-8(5)	-3(5)
C(10)	41(7)	70(8)	32(6)	-11(6)	-3(6)	-3(6)
C(11)	93(12)	55(7)	34(7)	-9(6)	2(8)	-26(9)
C(12)	57(10)	85(10)	52(10)	36(8)	-33(8)	-21(8)
C(13)	36(9)	103(13)	73(12)	-2(11)	-16(9)	-4(9)
C(14)	31(7)	79(9)	48(8)	2(7)	-2(7)	12(7)

The anisotropic temperature factor takes the form

$$\exp[-2\pi^2(h^2\underline{a}^{*2}\underline{U}_{11} + k^2\underline{b}^{*2}\underline{U}_{22} + l^2\underline{c}^{*2}\underline{U}_{33} + 2kl\underline{b}^*\underline{c}^*\underline{U}_{23} + 2lh\underline{c}^*\underline{a}^*\underline{U}_{31} + 2hk\underline{a}^*\underline{b}^*\underline{U}_{12})]$$

The common isotropic temperature factor for the hydrogen atoms refined to $63(13) \times 10^{-3}\AA^2$.

V. Bis(4-methylpyridine)hydrogen(I) tetraphenylborate.

(i) Structure Factors.

(ii) Thermal Parameters.

OBSERVED AND CALCULATED STRUCTURE FACTORS FOR GAMMAPICOLINE

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
4	0	0	1451	-1422	4	6	0	164	166	-9	3	1	129	-149	8	6	1	300	-276	-13	1	2	173	-179
6	0	0	844	-837	6	6	0	236	233	-7	3	1	169	-161	10	6	1	150	-160	-9	1	2	162	-166
8	0	0	295	-274	8	6	0	245	-236	-5	3	1	499	496	-9	7	1	148	117	-7	1	2	145	-153
10	0	0	226	215	1	7	0	89	-141	-3	3	1	614	652	-7	7	1	286	-273	-5	1	2	1367	-1325
12	0	0	270	-247	5	7	0	183	178	1	3	1	59	-106	-1	7	1	33	55	-3	1	2	669	-661
14	0	0	185	210	7	7	0	81	-73	3	3	1	259	-239	1	7	1	193	249	1	1	2	281	276
16	0	0	116	-150	9	7	0	122	112	7	3	1	479	-484	3	7	1	64	-61	3	1	2	1399	1356
3	1	0	552	556	11	7	0	91	80	13	3	1	169	-191	5	7	1	78	-64	5	1	2	641	575
5	1	0	748	-735	6	8	0	132	128	15	3	1	262	-302	7	7	1	362	-342	7	1	2	692	655
9	1	0	88	75	5	9	0	59	-43	-16	4	1	201	-217	9	7	1	185	-166	9	1	2	235	-228
11	1	0	184	-190	7	9	0	129	-114	-14	4	1	151	-143	11	7	1	135	129	15	1	2	217	-248
15	1	0	128	153	9	9	0	134	105	-12	4	1	201	-161	-6	8	1	84	-95	-16	2	2	161	-156
2	2	0	1224	1251	12	10	0	154	98	-8	4	1	246	-218	2	8	1	83	98	-12	2	2	339	292
4	2	0	1053	1013	-13	1	1	131	-159	-4	4	1	667	645	4	8	1	85	-81	-10	2	2	225	268
6	2	0	139	105	-11	1	1	152	-169	-2	4	1	96	144	6	8	1	291	-267	-8	2	2	586	-511
8	2	0	320	-317	-9	1	1	229	245	2	4	1	268	275	-9	9	1	87	-42	-2	2	2	686	707
10	2	0	219	-263	-7	1	1	185	161	6	4	1	106	-90	-7	9	1	117	101	0	2	2	62	45
14	2	0	207	-224	-5	1	1	943	-925	10	4	1	102	-133	-3	9	1	74	-84	2	2	2	1545	-1528
1	3	0	132	-154	-3	1	1	1112	1099	12	4	1	278	-232	5	9	1	96	-98	4	2	2	789	772
3	3	0	285	327	1	1	1	329	338	14	4	1	209	-234	-8	10	1	83	61	8	2	2	255	-267
5	3	0	413	404	3	1	1	155	149	16	4	1	145	144	-2	10	1	54	-81	10	2	2	206	206
7	3	0	95	106	5	1	1	233	-228	-13	5	1	292	-260	6	10	1	78	56	12	2	2	202	200
9	3	0	169	-165	7	1	1	359	334	-11	5	1	137	-151	8	10	1	98	-75	14	2	2	269	-284
11	3	0	101	-112	9	1	1	422	-453	-7	5	1	141	151	1	11	1	41	-74	16	2	2	253	-307
2	4	0	55	70	-14	2	1	183	-292	-5	5	1	136	136	-16	0	2	139	162	-15	3	2	174	163
4	4	0	201	-205	-10	2	1	275	-288	-3	5	1	229	248	-12	0	2	107	-90	-11	3	2	104	97
6	4	0	418	384	-8	2	1	621	-582	1	5	1	57	63	-10	0	2	83	-95	-9	3	2	92	-121
8	4	0	299	284	-6	2	1	559	545	3	5	1	82	-83	-8	0	2	186	-139	-7	3	2	1417	-1452
10	4	0	225	-248	-4	2	1	330	310	7	5	1	269	-210	-6	0	2	534	502	-5	3	2	262	-241
12	4	0	169	-172	2	2	1	1100	-1077	9	5	1	167	-155	-4	0	2	1391	-1404	-3	3	2	229	259
1	5	0	156	-218	6	2	1	67	68	-8	6	1	267	-258	-2	0	2	110	-103	1	3	2	512	528
3	5	0	223	258	8	2	1	324	320	-4	6	1	82	84	0	0	2	203	-245	3	3	2	103	-95
5	5	0	189	-164	10	2	1	257	-268	-2	6	1	254	346	2	0	2	329	-340	5	3	2	76	80
7	5	0	175	132	14	2	1	228	-235	0	6	1	53	127	8	0	2	576	549	7	3	2	218	-186
9	5	0	249	250	16	2	1	108	-127	2	6	1	276	-327	12	0	2	211	-217	9	3	2	228	259
11	5	0	128	-150	-15	3	1	402	-423	4	6	1	60	77	14	0	2	104	-110	11	3	2	142	132

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
15	3	2	137	-153	-10	8	2	208	-210	-7	3	3	795	-818	4	6	3	152	123	16	0	4	117	140
-14	4	2	177	179	-8	8	2	208	171	-5	3	3	359	-389	6	6	3	150	126	-13	1	4	347	364
-10	4	2	270	-283	-6	8	2	172	133	-3	3	3	393	406	8	6	3	274	266	-11	1	4	203	217
-6	4	2	111	93	-4	8	2	94	-111	-1	3	3	150	-165	10	6	3	241	-270	-9	1	4	362	384
-4	4	2	282	-271	-2	8	2	51	71	1	3	3	322	351	-13	7	3	159	-134	-7	1	4	151	124
-2	4	2	62	-71	0	8	2	65	-68	3	3	3	180	157	1	7	3	59	59	-5	1	4	81	-109
0	4	2	56	57	2	8	2	71	81	9	3	3	110	128	3	7	3	139	151	-3	1	4	243	-254
2	4	2	63	58	4	8	2	125	-123	11	3	3	160	151	5	7	3	217	206	-1	1	4	346	-360
4	4	2	195	-205	-9	9	2	111	-93	-14	4	3	349	335	9	7	3	229	-210	3	1	4	485	-488
6	4	2	112	127	-5	9	2	156	136	-12	4	3	144	115	-12	8	3	105	-109	5	1	4	477	-474
8	4	2	137	-137	-13	1	3	353	346	-10	4	3	378	-388	-10	8	3	86	-77	7	1	4	110	-95
12	4	2	222	-198	-11	1	3	236	-245	-8	4	3	573	-508	-6	8	3	214	209	9	1	4	103	92
-11	5	2	219	-234	-9	1	3	475	461	-6	4	3	81	110	-4	8	3	53	60	11	1	4	342	355
-9	5	2	206	-186	-7	1	3	398	-441	0	4	3	256	252	-2	8	3	51	-68	-14	2	4	149	159
-5	5	2	78	68	-5	1	3	648	-669	2	4	3	516	496	2	8	3	73	56	-12	2	4	200	175
-3	5	2	250	-270	-3	1	3	1291	-1324	4	4	3	113	-101	4	8	3	194	186	-10	2	4	299	324
-1	5	2	213	-303	-1	1	3	1153	1246	10	4	3	217	190	8	8	3	97	-75	-8	2	4	551	511
1	5	2	194	232	3	1	3	1359	-1337	14	4	3	131	-166	-5	9	3	126	132	-6	2	4	122	-89
3	5	2	215	197	5	1	3	138	128	-13	5	3	202	184	-6	10	3	122	-100	-4	2	4	715	-711
5	5	2	119	122	7	1	3	230	-226	-11	5	3	162	-177	-4	10	3	104	126	-2	2	4	415	-402
7	5	2	167	163	9	1	3	209	234	-9	5	3	252	-230	-2	10	3	52	72	0	2	4	1383	-1423
9	5	2	297	-281	-14	2	3	164	145	-5	5	3	87	-69	0	10	3	76	109	2	2	4	1003	1023
-11	5	2	308	-247	-8	2	3	199	-157	-3	5	3	245	-274	-3	11	3	48	78	4	2	4	210	262
15	5	2	128	119	-6	2	3	287	-282	-1	5	3	348	429	-16	0	4	173	-169	6	2	4	71	94
-10	6	2	118	-126	-4	2	3	352	-362	1	5	3	268	297	-14	0	4	722	711	12	2	4	252	253
-6	6	2	69	45	-2	2	3	271	271	3	5	3	228	-202	-12	0	4	434	392	14	2	4	154	167
0	6	2	139	169	0	2	3	153	137	5	5	3	201	-181	-10	0	4	69	68	16	2	4	124	121
2	6	2	143	-147	2	2	3	305	299	7	5	3	169	137	-8	0	4	247	-224	-13	3	4	135	-120
6	6	2	78	-79	4	2	3	139	-118	9	5	3	487	538	-6	0	4	175	205	-11	3	4	230	248
8	6	2	309	-304	6	2	3	178	183	11	5	3	155	-152	-2	0	4	180	-162	-7	3	4	339	324
-7	7	2	207	-187	8	2	3	214	216	13	5	3	145	-140	0	0	4	870	896	-5	3	4	315	316
-3	7	2	95	115	12	2	3	106	-81	-10	6	3	115	-115	2	0	4	1660	-1694	-3	3	4	361	-394
-1	7	2	81	106	-15	3	3	105	124	-4	6	3	119	-122	4	0	4	378	-372	-1	3	4	292	296
5	7	2	245	-220	-13	3	3	243	257	-2	6	3	141	160	6	0	4	657	-618	3	3	4	203	172
9	7	2	107	103	-11	3	3	150	162	0	6	3	154	-165	8	0	4	505	515	5	3	4	296	317
11	7	2	239	-204	-9	3	3	249	-224	2	6	3	133	149	10	0	4	841	776	7	3	4	102	-115

OBSERVED AND CALCULATED STRUCTURE FACTORS FOR GAMMAPICOLINE

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
11	3	4	142	123	7	7	4	181	196	6	2	5	188	182	-5	7	5	65	-62	-1	1	6	398	369
13	3	4	106	114	9	7	4	126	-133	8	2	5	232	256	-1	7	5	70	56	1	1	6	77	-27
-14	4	4	126	120	-10	8	4	91	73	10	2	5	141	-140	3	7	5	152	-150	3	1	6	636	-572
-12	4	4	248	216	-8	8	4	91	77	-15	3	5	145	-126	5	7	5	183	163	7	1	6	234	-258
-8	4	4	89	103	-6	8	4	368	-279	-11	3	5	259	256	-6	8	5	120	-101	-12	2	6	327	-298
-4	4	4	105	-98	2	8	4	134	151	-9	3	5	225	227	-2	8	5	63	-60	-10	2	6	110	-129
0	4	4	329	-315	4	8	4	183	152	-7	3	5	563	-607	2	8	5	87	62	-8	2	6	106	94
2	4	4	491	-476	8	8	4	147	-134	-3	3	5	247	257	10	8	5	121	-91	-6	2	6	332	-356
4	4	4	428	420	-11	9	4	87	75	-1	3	5	553	561	-9	9	5	149	-137	-4	2	6	160	172
6	4	4	139	120	-7	9	4	146	-145	1	3	5	1137	-1197	-5	9	5	153	-148	-2	2	6	73	-53
8	4	4	343	369	-1	9	4	47	53	3	3	5	500	-526	-3	9	5	98	-87	0	2	6	322	-287
10	4	4	167	169	1	9	4	74	79	5	3	5	417	416	1	9	5	63	81	2	2	6	139	-152
12	4	4	118	-117	-4	10	4	71	-84	9	3	5	213	193	3	9	5	183	155	-9	3	6	74	84
-9	5	4	153	-137	-2	10	4	54	71	11	3	5	111	-103	-2	10	5	65	-60	-5	3	6	199	257
-7	5	4	101	-111	0	10	4	51	46	15	3	5	128	-167	0	10	5	53	49	-3	3	6	88	119
-5	5	4	302	316	-15	1	5	282	288	-12	4	5	193	173	4	10	5	70	73	-1	3	6	95	104
-3	5	4	150	152	-13	1	5	108	-125	-10	4	5	358	360	-16	0	6	187	203	1	3	6	539	-591
-1	5	4	181	-197	-11	1	5	121	-123	-6	4	5	369	-391	-14	0	6	574	-568	3	3	6	957	-894
3	5	4	69	43	-9	1	5	161	-127	-4	4	5	185	215	-12	0	6	485	-456	5	3	6	178	179
7	5	4	544	537	-7	1	5	101	94	-2	4	5	158	-190	-10	0	6	304	307	-10	4	6	98	85
9	5	4	121	146	-5	1	5	496	520	2	4	5	106	112	-8	0	6	148	-140	-6	4	6	433	-495
11	5	4	152	-139	-3	1	5	214	-234	4	4	5	350	-352	-6	0	6	350	350	-4	4	6	82	82
-16	6	4	226	236	1	1	5	225	249	8	4	5	160	-197	-4	0	6	937	-923	0	4	6	456	-455
-10	6	4	137	-119	3	1	5	407	-402	10	4	5	167	148	-2	0	6	206	220	2	4	6	193	179
-2	6	4	63	63	5	1	5	401	-377	-13	5	5	131	84	0	0	6	657	-653	4	4	6	214	-206
0	6	4	177	-177	7	1	5	323	326	-5	5	5	152	-160	2	0	6	210	215	6	4	6	123	125
2	6	4	135	-123	9	1	5	150	138	-3	5	5	133	-111	4	0	6	269	246	8	4	6	126	-99
4	6	4	116	119	-16	2	5	173	177	-1	5	5	93	-89	6	0	6	113	-96	10	4	6	130	-127
6	6	4	313	271	-14	2	5	216	-217	5	5	5	142	-124	10	0	6	352	-345	-13	5	6	289	266
8	6	4	191	190	-8	2	5	455	435	-8	6	5	163	152	16	0	6	177	-231	-7	5	6	206	213
10	6	4	199	165	-6	2	5	544	585	-4	6	5	208	-228	-15	1	6	295	-281	-5	5	6	120	-137
12	6	4	140	-113	-4	2	5	533	-565	0	6	5	115	-107	-13	1	6	195	-153	-1	5	6	147	-171
-5	7	4	103	121	-2	2	5	616	646	6	6	5	110	124	-9	1	6	324	328	1	5	6	131	82
-1	7	4	78	68	0	2	5	986	-1084	8	6	5	200	-199	-7	1	6	294	265	5	5	6	292	-234
1	7	4	122	-142	2	2	5	736	-662	10	6	5	138	-121	-5	1	6	394	-390	7	5	6	202	-210
5	7	4	344	337	4	2	5	102	120	-7	7	5	277	-266	-3	1	6	351	-337	11	5	6	283	290

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
-14	6	6	188	175	7	1	7	242	-226	5	5	7	106	125	-9	1	8	135	132	8	4	8	128	111
-12	6	6	146	147	9	1	7	248	227	9	5	7	176	157	-7	1	8	127	-98	-7	5	8	118	142
-2	6	6	178	-193	-16	2	7	367	-218	11	5	7	181	-184	-5	1	8	71	-16	-5	5	8	301	317
0	6	6	91	91	-12	2	7	435	418	-12	6	7	142	107	-1	1	8	299	-287	-3	5	8	221	266
2	6	6	118	-115	-8	2	7	73	88	8	6	7	152	107	1	1	8	115	106	-1	5	8	85	-99
4	6	6	188	-189	-6	2	7	656	-631	-15	7	7	162	120	3	1	8	91	76	3	5	8	201	-185
6	6	6	293	-305	-4	2	7	172	-166	-5	7	7	76	30	7	1	8	190	-179	-12	6	8	130	-119
8	6	6	215	233	-2	2	7	281	295	5	7	7	105	101	9	1	8	100	97	-6	6	8	360	384
10	6	6	202	197	0	2	7	476	496	7	7	7	86	79	11	1	8	198	242	-4	6	8	333	335
-11	7	6	113	133	2	2	7	721	663	-8	8	7	120	113	-12	2	8	186	167	-9	7	8	95	116
-7	7	6	98	80	4	2	7	559	-572	-6	8	7	99	117	-10	2	8	154	179	-7	7	8	224	219
-3	7	6	93	-108	6	2	7	126	-119	-4	8	7	85	-91	-8	2	8	100	-105	-5	7	8	185	-166
1	7	6	229	-214	-11	3	7	88	91	-2	8	7	85	-104	-6	2	8	313	301	-3	7	8	98	107
3	7	6	161	-147	-9	3	7	137	-166	0	8	7	94	-116	-4	2	8	289	290	-1	7	8	156	165
7	7	6	140	133	-7	3	7	104	-123	4	8	7	133	127	-2	2	8	210	-163	9	7	8	119	-118
-6	8	6	125	-112	-5	3	7	100	115	-9	9	7	87	72	2	2	8	980	927	-6	8	8	131	-123
-4	8	6	74	68	-3	3	7	145	157	-1	9	7	141	-144	4	2	8	415	-424	-4	8	8	187	181
0	8	6	193	-187	-1	3	7	88	132	5	9	7	97	84	6	2	8	258	-224	-2	8	8	152	156
2	8	6	80	-99	1	3	7	354	338	-2	10	7	81	-52	8	2	8	220	-208	0	8	8	192	180
-9	9	6	82	-66	3	3	7	443	429	6	10	7	111	75	10	2	8	131	122	2	8	8	103	-102
-7	9	6	166	158	5	3	7	301	-315	-5	11	7	97	-82	12	2	8	291	372	4	8	8	128	-116
-5	9	6	118	111	9	3	7	103	-101	-16	0	8	115	-109	-11	3	8	181	163	-1	9	8	83	73
-3	9	6	135	-138	11	3	7	109	96	-14	0	8	105	-113	-7	3	8	190	204	2	10	8	84	71
-1	9	6	89	-100	-6	4	7	87	99	-12	0	8	113	94	-1	3	8	488	462	-15	1	9	108	97
1	9	6	154	-161	-4	4	7	109	92	-10	0	8	139	165	1	3	8	670	-666	-13	1	9	459	-451
-2	10	6	110	-131	-2	4	7	63	-63	-8	0	8	340	363	5	3	8	90	113	-9	1	9	178	205
6	10	6	99	-63	0	4	7	148	111	-6	0	8	315	280	7	3	8	140	-133	-7	1	9	287	265
-13	1	7	417	372	2	4	7	275	239	-4	0	8	124	-103	9	3	8	237	257	-3	1	9	634	-648
-11	1	7	87	-88	6	4	7	168	-186	-2	0	8	124	-124	-10	4	8	102	89	-1	1	9	251	254
-7	1	7	592	-580	8	4	7	197	-182	0	0	8	119	-141	-6	4	8	157	141	3	1	9	210	-221
-5	1	7	79	66	10	4	7	309	306	2	0	8	313	321	-4	4	8	479	476	5	1	9	134	143
-3	1	7	237	242	-9	5	7	135	-155	4	0	8	196	179	-2	4	8	253	291	7	1	9	209	197
-1	1	7	731	735	-7	5	7	109	112	6	0	8	159	-139	0	4	8	77	97	-10	2	9	89	101
1	1	7	236	283	-5	5	7	327	380	14	0	8	142	177	2	4	8	483	-464	-8	2	9	237	256
3	1	7	507	-482	1	5	7	261	-263	-13	1	8	464	453	4	4	8	72	66	-6	2	9	229	-208
5	1	7	79	-65	3	5	7	295	282	-11	1	8	107	98	6	4	8	83	86	-4	2	9	431	-432

H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC	H	K	L	10FO	10FC
-2	2	9	95	-125	-4	0	10	394	-417	-4	4	10	145	-153	-4	2	11	171	204	-16	0	12	98	-114
0	2	9	97	63	0	0	10	273	281	-2	4	10	75	70	-2	2	11	131	153	-12	0	12	369	365
6	2	9	111	95	2	0	10	86	-85	0	4	10	180	189	4	2	11	234	-214	-10	0	12	253	-237
8	2	9	202	195	4	0	10	515	-484	2	4	10	181	203	6	2	11	134	-126	-8	0	12	229	-216
-15	3	9	108	105	6	0	10	154	-139	4	4	10	307	-292	12	2	11	126	168	-6	0	12	381	-346
-3	3	9	173	-162	8	0	10	132	-93	8	4	10	175	-155	-15	3	11	193	-207	-2	0	12	151	166
1	3	9	298	-303	-15	1	10	96	57	-11	5	10	113	-101	-11	3	11	489	482	2	0	12	602	566
9	3	9	201	217	-13	1	10	299	-309	-9	5	10	195	-232	-5	3	11	107	-114	4	0	12	539	506
-8	4	9	198	-212	-11	1	10	449	-443	-7	5	10	206	-232	-3	3	11	83	79	6	0	12	204	-185
-6	4	9	279	-327	-9	1	10	76	79	-5	5	10	131	-159	-1	3	11	80	-73	8	0	12	138	-159
-4	4	9	199	211	-7	1	10	89	98	-3	5	10	228	228	1	3	11	150	139	-9	1	12	94	-76
0	4	9	175	182	-5	1	10	334	334	1	5	10	140	141	3	3	11	202	188	-7	1	12	233	-213
2	4	9	420	-434	1	1	10	140	-102	3	5	10	91	108	5	3	11	383	-364	-3	1	12	65	79
4	4	9	179	-244	5	1	10	117	-114	5	5	10	266	-231	7	3	11	255	-262	1	1	12	417	369
6	4	9	276	257	7	1	10	120	-152	13	5	10	132	-132	11	3	11	304	381	3	1	12	586	534
8	4	9	196	177	11	1	10	171	-218	-10	6	10	146	-115	13	3	11	99	144	9	1	12	160	-198
10	4	9	195	213	-16	2	10	121	-139	-8	6	10	278	-324	-16	4	11	137	-168	-16	2	12	128	136
-5	5	9	293	-291	-12	2	10	150	-127	2	6	10	89	-99	-14	4	11	115	-100	-6	2	12	180	-174
5	5	9	129	-109	-8	2	10	183	203	4	6	10	130	120	-12	4	11	112	100	-4	2	12	117	-135
7	5	9	228	212	-4	2	10	184	193	-9	7	10	120	-121	-10	4	11	386	357	-2	2	12	191	219
9	5	9	137	-106	0	2	10	146	144	-7	7	10	155	-164	-4	4	11	84	100	0	2	12	352	310
-8	6	9	141	150	2	2	10	182	-189	-5	7	10	103	-88	4	4	11	126	119	6	2	12	275	297
-6	6	9	175	-154	4	2	10	211	-200	-3	7	10	98	86	6	4	11	319	-309	-15	3	12	106	93
-4	6	9	204	-222	6	2	10	230	-239	-8	8	10	83	-77	8	4	11	124	-133	-13	3	12	216	-222
-7	7	9	96	109	10	2	10	127	-127	-2	8	10	85	-78	10	4	11	133	129	-3	3	12	80	131
-5	7	9	111	114	-15	3	10	105	-135	4	8	10	92	85	-11	5	11	127	111	-1	3	12	159	140
-3	7	9	190	-193	-13	3	10	94	-109	3	9	10	104	119	-9	5	11	224	200	3	3	12	212	207
-1	7	9	106	-87	-11	3	10	121	-129	-11	1	11	130	-102	-7	5	11	80	72	5	3	12	131	134
-3	9	9	117	94	-5	3	10	207	-233	-9	1	11	116	86	-6	6	11	134	158	7	3	12	131	121
1	9	9	94	-84	-3	3	10	75	91	-7	1	11	103	98	-4	6	11	291	336	-14	4	12	154	161
-6	10	9	98	79	-1	3	10	131	144	-1	1	11	141	151	2	6	11	223	-190	-8	4	12	113	-105
-5	11	9	95	60	3	3	10	85	89	1	1	11	82	-79	-7	7	11	101	105	-4	4	12	121	136
-14	0	10	370	-363	9	3	10	203	-220	3	1	11	178	-185	-5	7	11	247	250	2	4	12	125	-115
-12	0	10	166	-170	11	3	10	164	-211	-12	2	11	289	282	-1	7	11	107	-92	4	4	12	195	188
-8	0	10	219	248	-8	4	10	68	-72	-8	2	11	75	91	-4	8	11	79	43	-5	5	12	189	213
-6	0	10	739	790	-6	4	10	79	-94	-6	2	11	135	-151	-2	8	11	101	101	-3	5	12	91	107

H	K	L	10F$_o$	10FC
-1	5	12	91	-107
3	5	12	172	-147
-4	6	12	183	-198
-2	6	12	79	87
-9	7	12	120	91
-5	7	12	89	-99
-3	7	12	102	120
-6	8	12	90	92
-7	1	13	173	122
-5	1	13	117	-79
-3	1	13	313	-302
-1	1	13	316	309
1	1	13	317	294
3	1	13	155	-146
5	1	13	133	-155
-16	2	13	114	-101
-12	2	13	215	-219
0	2	13	283	272
4	2	13	142	-123
6	2	13	114	-98
-13	3	13	156	-162
-11	3	13	182	-207
-3	3	13	180	178
1	3	13	109	102
-12	4	13	226	-223
-10	4	13	163	-170
-6	4	13	206	187
4	4	13	128	128
10	4	13	122	-123
-11	5	13	113	-114
-9	5	13	175	-157
-5	5	13	180	-162
-1	5	13	115	104
1	5	13	222	214
5	5	13	158	-176
0	6	13	109	90
2	6	13	140	126
4	6	13	121	-79
6	6	13	141	-145
-1	7	13	156	117
-10	0	14	141	-155
-4	0	14	117	-122
-2	0	14	401	-394
0	0	14	409	-395
4	0	14	198	224
-15	1	14	124	134
-3	1	14	472	-465
-1	1	14	378	-369
7	1	14	115	152
-16	2	14	93	107
-14	2	14	219	235
-12	2	14	135	-126
-10	2	14	160	-156
-6	2	14	174	-156
-4	2	14	168	-160
-2	2	14	268	-239
2	2	14	131	-112
6	2	14	111	107
8	2	14	141	164
-13	3	14	142	159
-9	3	14	139	-110
-5	3	14	287	-291
-3	3	14	136	-139
-12	4	14	181	183
-8	4	14	196	-192
-6	4	14	230	-192
-4	4	14	101	-110
-2	4	14	163	138
-11	5	14	134	119
-7	5	14	181	-172
-4	6	14	117	-100
-3	1	15	176	-169
3	1	15	213	226
-4	2	15	247	-250
2	2	15	204	218
4	2	15	105	115
-11	3	15	172	202
-7	3	15	152	-175
-5	3	15	355	-355
-1	3	15	137	151
-10	4	15	219	221
-8	4	15	175	-177
-6	4	15	221	-215
-4	4	15	194	-195
4	4	15	132	155
-7	5	15	186	-187
-2	6	15	151	-134
6	6	15	123	128
-10	0	16	152	180
-8	0	16	99	129
-4	0	16	238	234
0	0	16	249	-249
4	0	16	127	156
-11	1	16	194	217
-7	1	16	138	128
-10	2	16	184	191
-8	2	16	203	226
-6	2	16	100	124
-9	3	16	171	174
-7	3	16	119	123
-10	4	16	121	115
-4	4	16	141	159
-12	6	16	119	96
8	6	16	138	147

Table B. Anisotropic Thermal Parameters ($Å^2 \times 10^3$) and Equivalent Isotropic Thermal Parameters ($Å^2 \times 10^3$)

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{13}	U_{12}	U_{iso}*
B(1)	29(5)	64(7)	29(5)	0(0)	6(4)	0(0)	37(6)
C(11)	14(3)	92(6)	26(3)	8(3)	1(2)	5(3)	32(4)
C(12)	41(4)	89(6)	55(4)	1(4)	1(3)	-14(4)	57(3)
C(13)	52(5)	94(7)	75(6)	17(5)	-13(4)	-23(5)	67(6)
C(14)	34(5)	135(10)	86(7)	47(7)	3(5)	-27(6)	62(7)
C(15)	29(4)	129(9)	58(5)	36(6)	6(3)	-8(5)	55(6)
C(16)	25(3)	102(6)	41(4)	21(4)	5(3)	5(4)	45(4)
C(21)	16(3)	85(5)	26(3)	1(3)	7(2)	7(3)	31(4)
C(22)	26(3)	83(5)	27(4)	0(4)	5(2)	10(3)	38(4)
C(23)	30(3)	108(7)	38(4)	-7(4)	5(3)	4(4)	49(5)
C(24)	28(3)	129(8)	40(5)	-25(5)	-1(3)	-4(4)	50(5)
C(25)	29(3)	96(7)	64(5)	-17(5)	11(3)	-18(4)	53(5)
C(26)	31(3)	88(6)	40(4)	-7(4)	13(3)	-10(4)	45(4)
N(1)	52(4)	121(6)	57(4)	14(4)	6(3)	6(4)	70(4)
C(1)	59(5)	88(6)	63(5)	12(5)	8(4)	24(5)	66(5)
C(2)	67(5)	87(6)	61(5)	7(5)	14(4)	7(5)	70(5)
C(3)	54(4)	105(7)	68(5)	18(5)	9(4)	1(5)	71(5)
C(4)	48(5)	132(10)	132(8)	60(8)	14(5)	25(6)	84(8)
C(5)	77(6)	102(8)	115(8)	48(6)	-4(6)	15(6)	89(7)
C(6)	60(5)	207(12)	127(8)	64(8)	37(5)	3(6)	102(8)

* Equivalent isotropic thermal parameters are defined as the geometric mean of the diagonal components of the diagonalised matrix of U_{ij}.

Isotropic thermal parameters refined to the values /$Å^2 \times 10^3$:

H(7)	120(40)
Phenyl H	75(7)
Pyridine H	77(6)
Methyl H	277(40)

VI. Bis(triphenylarsineoxide)hydrogen(I) tetrafluoroborate.

(i) Structure Factors.

(ii) Thermal Parameters.

OBSERVED AND CALCULATED STRUCTURE FACTORS FOR HHARSENIC

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
2	0	0	46	44	2	4	0	35	-41	11	7	0	26	20	4	13	0	56	-59	4	1	1	17	-19
3	0	0	149	159	3	4	0	68	-70	1	8	0	37	59	6	13	0	21	20	5	1	1	20	19
4	0	0	10	19	4	4	0	27	-30	2	8	0	20	24	7	13	0	39	-35	6	1	1	22	-22
5	0	0	52	54	5	4	0	72	-68	3	8	0	51	-51	8	13	0	21	21	7	1	1	49	-49
6	0	0	100	109	6	4	0	62	-66	5	8	0	56	46	10	13	0	36	-27	9	1	1	34	-36
7	0	0	69	68	7	4	0	28	-26	6	8	0	37	-38	1	14	0	9	19	10	1	1	58	-57
8	0	0	16	15	8	4	0	32	-29	9	8	0	32	-34	2	14	0	9	17	-10	2	1	79	-71
9	0	0	83	94	9	4	0	60	-62	1	9	0	6	8	3	14	0	12	-15	-7	2	1	24	12
10	0	0	79	78	10	4	0	59	-52	2	9	0	7	6	4	14	0	18	18	-5	2	1	91	90
12	0	0	44	46	12	4	0	30	-32	3	9	0	15	12	5	14	0	33	35	-4	2	1	95	-97
13	0	0	39	41	1	5	0	6	7	4	9	0	38	34	8	14	0	23	21	-3	2	1	24	17
16	0	0	22	29	2	5	0	58	-65	6	9	0	29	-32	2	15	0	8	-12	-2	2	1	206	201
2	1	0	116	123	3	5	0	39	-39	8	9	0	36	-37	1	16	0	9	-21	1	2	1	133	145
4	1	0	10	-20	4	5	0	23	25	1	10	0	20	-36	2	16	0	11	-21	2	2	1	106	-106
5	1	0	86	82	5	5	0	63	-58	2	10	0	48	-66	3	16	0	15	-20	3	2	1	73	-78
7	1	0	101	-95	7	5	0	60	56	3	10	0	62	-73	4	16	0	15	-16	4	2	1	37	37
8	1	0	17	18	10	5	0	39	35	4	10	0	29	-35	6	16	0	25	-28	6	2	1	20	-22
9	1	0	25	25	1	6	0	9	-12	5	10	0	45	-47	7	16	0	31	-32	7	2	1	40	42
10	1	0	49	-43	2	6	0	58	65	6	10	0	50	-49	8	16	0	19	-18	8	2	1	19	19
3	2	0	87	-85	3	6	0	148	161	7	10	0	40	-43	9	16	0	22	-20	9	2	1	31	-34
4	2	0	55	-54	4	6	0	45	47	8	10	0	16	-11	1	17	0	8	16	10	2	1	52	40
6	2	0	95	-100	5	6	0	33	34	10	10	0	36	-31	4	17	0	20	24	11	2	1	33	33
7	2	0	49	-45	6	6	0	128	133	12	10	0	34	-32	3	18	0	18	24	-15	3	1	34	43
8	2	0	25	29	7	6	0	65	66	1	11	0	13	23	6	18	0	25	23	-12	3	1	71	81
9	2	0	31	-34	9	6	0	69	76	2	11	0	27	-35	1	19	0	7	-16	-10	3	1	43	40
10	2	0	34	-31	10	6	0	78	72	4	11	0	43	51	-12	1	1	42	-44	-9	3	1	95	112
12	2	0	22	-19	12	6	0	56	56	7	11	0	34	36	-10	1	1	41	-32	-6	3	1	134	117
1	3	0	28	34	13	6	0	31	31	3	12	0	56	68	-9	1	1	51	-62	-5	3	1	74	73
2	3	0	72	80	1	7	0	14	-27	4	12	0	22	25	-6	1	1	60	-56	-4	3	1	42	43
3	3	0	30	-29	2	7	0	89	106	5	12	0	13	-16	-5	1	1	36	-31	-3	3	1	107	114
4	3	0	37	34	3	7	0	46	50	6	12	0	63	64	-4	1	1	16	-17	-2	3	1	189	191
5	3	0	29	26	4	7	0	27	-35	7	12	0	34	31	-3	1	1	51	40	-1	3	1	10	8
7	3	0	20	16	5	7	0	33	35	9	12	0	41	41	-2	1	1	143	-137	1	3	1	60	61
8	3	0	28	28	7	7	0	54	-54	12	12	0	38	33	1	1	1	126	-138	2	3	1	30	30
11	3	0	24	-22	9	7	0	28	29	1	13	0	17	-37	2	1	1	59	-61	3	3	1	109	120
1	4	0	22	-23	10	7	0	43	-35	2	13	0	12	24	3	1	1	146	-162	4	3	1	79	75

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
6	3	1	37	36	5	5	1	24	26	7	7	1	69	-62	13	9	1	37	31	-10	13	1	40	-32
7	3	1	113	113	6	5	1	19	-21	8	7	1	29	-25	-10	10	1	42	34	-9	13	1	26	-27
9	3	1	66	67	7	5	1	25	-30	9	7	1	28	-25	-8	10	1	32	-27	-8	13	1	26	-23
10	3	1	105	96	10	5	1	37	-27	10	7	1	55	-48	-7	10	1	45	48	-7	13	1	29	-32
13	3	1	49	48	11	5	1	32	26	-10	8	1	49	-41	-5	10	1	46	-47	-6	13	1	36	-43
-11	4	1	36	-31	13	5	1	27	-27	-8	8	1	21	18	-4	10	1	67	76	-5	13	1	52	-51
-10	4	1	44	41	-10	6	1	27	15	-7	8	1	45	-44	-3	10	1	19	21	-4	13	1	38	-39
-8	4	1	35	-33	-8	6	1	18	17	-5	8	1	52	54	-2	10	1	10	-13	-3	13	1	47	-58
-7	4	1	64	68	-7	6	1	51	-49	-4	8	1	55	-64	0	10	1	7	16	-2	13	1	33	-52
-5	4	1	83	-86	-6	6	1	19	-14	-2	8	1	87	105	1	10	1	48	-78	-1	13	1	17	-30
-4	4	1	120	127	-5	6	1	28	-24	0	8	1	24	-43	3	10	1	33	42	0	13	1	16	-39
-2	4	1	116	-123	-4	6	1	21	-18	1	8	1	40	61	4	10	1	56	-56	1	13	1	22	-39
-1	4	1	48	54	-2	6	1	65	-66	3	8	1	39	-43	5	10	1	21	-26	2	13	1	30	-43
0	4	1	31	46	-1	6	1	83	93	4	8	1	60	59	6	10	1	43	47	3	13	1	40	-53
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7	4	1	67	-69	-12	7	1	35	-34	-9	9	1	62	70	-5	11	1	37	39	10	13	1	36	-29
8	4	1	38	-39	-10	7	1	29	-30	-7	9	1	28	27	-4	11	1	34	37	-8	14	1	23	16
9	4	1	30	32	-9	7	1	57	-65	-6	9	1	117	112	-2	11	1	25	28	-5	14	1	16	18
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-9	5	1	42	-46	-6	7	1	47	-43	-3	9	1	83	92	5	11	1	60	61	4	14	1	15	20
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-4	5	1	18	19	-4	7	1	68	-65	-1	9	1	25	-36	8	11	1	35	29	-9	15	1	34	35
-3	5	1	24	-25	-3	7	1	108	-106	1	9	1	66	100	11	11	1	29	20	-6	15	1	38	41
-2	5	1	32	-35	-2	7	1	81	-92	3	9	1	100	116	-7	12	1	22	-25	-3	15	1	26	34
-1	5	1	85	95	-1	7	1	28	-38	4	9	1	65	66	-4	12	1	25	-26	-1	15	1	10	-15
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2	5	1	12	16	4	7	1	77	-73	9	9	1	29	27	2	12	1	13	16	3	15	1	24	30
3	5	1	77	-78	5	7	1	53	-52	10	9	1	68	53	6	12	1	16	-15	4	15	1	24	25
4	5	1	53	-53	6	7	1	36	-38	12	9	1	30	30	-12	13	1	41	-30	6	15	1	18	15

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
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10	15	1	40	28	-9	1	2	40	38	-4	3	2	16	17	-3	5	2	44	-43	-8	7	2	44	-47
-2	16	1	16	-22	-8	1	2	53	-55	-1	3	2	61	53	-2	5	2	101	110	-6	7	2	68	59
0	16	1	6	12	-7	1	2	22	-20	1	3	2	17	16	-1	5	2	51	-55	-5	7	2	91	-91
1	16	1	12	-22	-6	1	2	50	46	2	3	2	174	-176	0	5	2	70	-80	-4	7	2	35	-29
3	16	1	12	17	-5	1	2	105	-102	3	3	2	20	-19	1	5	2	36	45	-3	7	2	52	52
6	16	1	25	22	-4	1	2	56	45	5	3	2	43	40	2	5	2	33	39	-2	7	2	53	-55
-7	17	1	26	25	-3	1	2	189	180	6	3	2	29	28	3	5	2	67	-64	-1	7	2	50	-54
-4	17	1	21	26	-2	1	2	119	-124	-12	4	2	53	-56	4	5	2	52	51	0	7	2	29	44
-2	17	1	13	21	-1	1	2	20	-20	-11	4	2	42	-41	5	5	2	50	53	1	7	2	28	-37
-1	17	1	17	34	0	1	2	90	94	-10	4	2	45	-40	6	5	2	25	-26	3	7	2	86	92
2	17	1	21	29	1	1	2	83	-91	-9	4	2	102	-84	7	5	2	36	34	4	7	2	41	-43
5	17	1	24	25	2	1	2	270	-310	-8	4	2	45	-49	8	5	2	57	59	5	7	2	32	-41
-5	19	1	17	-14	3	1	2	83	83	-6	4	2	33	-27	11	5	2	34	31	6	7	2	45	44
-3	19	1	15	-24	4	1	2	71	-73	-5	4	2	101	-95	-15	6	2	31	38	8	7	2	35	-34
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-15	0	2	32	41	7	1	2	48	-46	-3	4	2	139	-122	-11	6	2	37	33	-6	8	2	33	-33
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-9	0	2	155	140	-12	2	2	28	-31	0	4	2	109	-115	-6	6	2	170	153	-1	8	2	65	75
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-5	0	2	133	131	-6	2	2	100	-89	3	4	2	111	-111	-3	6	2	102	104	2	8	2	45	52
-3	0	2	318	308	-5	2	2	52	-45	4	4	2	49	-44	-2	6	2	114	125	3	8	2	19	-20
-2	0	2	200	209	-3	2	2	124	-113	5	4	2	31	-27	-1	6	2	49	-54	4	8	2	77	-74
0	0	2	173	200	-2	2	2	149	-145	6	4	2	58	-59	0	6	2	62	80	5	8	2	37	35
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3	0	2	183	190	0	2	2	139	-147	8	4	2	28	-31	3	6	2	132	133	-11	9	2	39	12
4	0	2	216	229	1	2	2	91	-94	9	4	2	41	-42	4	6	2	124	126	-5	9	2	27	25
5	0	2	25	23	2	2	2	36	34	10	4	2	98	-86	5	6	2	25	24	-4	9	2	18	-17
6	0	2	85	92	3	2	2	80	-77	-11	5	2	45	40	6	6	2	53	53	-3	9	2	35	-35
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13	0	2	37	38	-7	3	2	26	-18	-6	5	2	48	-48	10	6	2	95	82	5	9	2	52	-50
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H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
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-12	10	2	34	-32	-9	12	2	40	42	-6	16	2	21	-20	2	1	3	65	-68	-2	3	3	158	148
-11	10	2	30	-21	-8	12	2	21	21	-5	16	2	26	-27	3	1	3	98	94	-1	3	3	36	-33
-10	10	2	27	-29	-6	12	2	49	49	-3	16	2	21	-30	4	1	3	70	-77	1	3	3	198	204
-9	10	2	28	-30	-5	12	2	53	56	-2	16	2	25	-40	5	1	3	49	-53	2	3	3	62	62
-7	10	2	39	-35	-4	12	2	21	-17	-1	16	2	16	-22	6	1	3	37	-36	3	3	3	47	-44
-6	10	2	69	-64	-3	12	2	42	52	0	16	2	19	-31	7	1	3	73	-78	4	3	3	140	154
-5	10	2	58	-60	-2	12	2	68	89	1	16	2	12	-16	8	1	3	28	-32	5	3	3	33	37
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6	10	2	31	-24	-3	13	2	42	54	0	18	2	10	21	0	2	3	23	17	-7	4	3	35	-29
7	10	2	45	-40	-2	13	2	19	-22	1	18	2	13	20	1	2	3	49	48	-6	4	3	58	56
8	10	2	49	-22	0	13	2	17	26	4	18	2	19	24	2	2	3	170	175	-5	4	3	65	-67
9	10	2	26	-25	1	13	2	31	-50	7	18	2	22	17	3	2	3	114	107	-4	4	3	52	-47
10	10	2	42	-32	2	13	2	13	-27	0	19	2	10	16	4	2	3	16	17	-3	4	3	58	53
-5	11	2	45	46	3	13	2	32	37	3	19	2	15	19	5	2	3	134	141	-2	4	3	81	-79
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8	11	2	30	26	-6	15	2	26	-22	0	1	3	37	35	-4	3	3	18	16	-4	5	3	20	19

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
-2	5	3	107	-100	7	7	3	75	-69	-6	10	3	55	54	-6	13	3	38	-43	-1	17	3	24	32
-1	5	3	58	54	8	7	3	49	-49	-5	10	3	29	-30	-5	13	3	32	-37	0	17	3	21	31
0	5	3	29	-25	10	7	3	48	-40	-4	10	3	36	-37	-4	13	3	44	-43	3	17	3	15	18
2	5	3	27	26	-9	8	3	31	-22	-3	10	3	50	59	-3	13	3	27	-28	3	18	3	14	-20
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4	5	3	90	-89	-7	8	3	19	21	-1	10	3	29	-35	-1	13	3	28	-33	0	19	3	15	-24
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-8	6	3	20	-21	-4	8	3	42	43	3	10	3	40	46	2	13	3	26	-28	-11	0	4	82	89
-6	6	3	23	-20	-3	8	3	68	-65	5	10	3	70	-72	3	13	3	33	-39	-9	0	4	52	48
-5	6	3	15	9	-2	8	3	59	64	6	10	3	18	17	4	13	3	40	-45	-8	0	4	159	170
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5	6	3	24	21	8	8	3	72	75	-2	11	3	55	-58	0	14	3	17	-22	0	0	4	47	47
7	6	3	22	-19	-12	9	3	38	42	-1	11	3	21	28	5	14	3	28	32	1	0	4	111	124
-12	7	3	39	-47	-11	9	3	36	29	1	11	3	11	-14	8	14	3	21	17	2	0	4	34	35
-11	7	3	37	-35	-9	9	3	73	58	2	11	3	24	29	-8	15	3	21	21	3	0	4	44	-46
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-6	7	3	62	-62	-5	9	3	102	112	5	11	3	25	18	-2	15	3	44	58	6	0	4	24	-24
-5	7	3	75	-76	-3	9	3	49	45	6	11	3	25	28	0	15	3	14	24	7	0	4	128	133
-4	7	3	40	-41	-2	9	3	152	159	9	11	3	31	24	1	15	3	23	29	8	0	4	96	111
-3	7	3	90	-85	0	9	3	36	45	-6	12	3	25	-24	4	15	3	30	34	10	0	4	49	43
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1	7	3	103	-115	5	9	3	36	35	4	12	3	14	-11	-1	16	3	13	-14	-12	1	4	38	43
2	7	3	33	-35	7	9	3	86	75	7	12	3	26	-23	0	16	3	16	21	-11	1	4	38	-37
3	7	3	18	-14	8	9	3	31	31	-11	13	3	33	-25	2	16	3	16	-25	-10	1	4	33	-26
4	7	3	63	-68	10	9	3	42	30	-9	13	3	39	-31	3	16	3	16	19	-9	1	4	52	49
5	7	3	51	-51	-9	10	3	34	29	-8	13	3	30	-32	-7	17	3	24	20	-8	1	4	34	-37
6	7	3	37	-35	-8	10	3	32	-33	-7	13	3	32	-27	-4	17	3	22	25	-7	1	4	100	-100

OBSERVED AND CALCULATED STRUCTURE FACTORS FOR HHARSENIC

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
-6	1	4	48	51	4	3	4	30	-27	8	5	4	59	65	8	7	4	28	-30	5	10	4	50	-51
-5	1	4	31	-27	8	3	4	27	-34	11	5	4	32	27	-11	8	4	28	-25	6	10	4	29	-24
-4	1	4	47	-54	-14	4	4	29	-40	12	5	4	34	30	-7	8	4	23	21	7	10	4	27	-22
-3	1	4	153	136	-12	4	4	36	-38	-14	6	4	40	54	-5	8	4	80	-71	8	10	4	25	-27
-1	1	4	265	-267	-11	4	4	48	-50	-12	6	4	34	43	-3	8	4	56	50	-7	11	4	46	45
1	1	4	18	19	-10	4	4	37	-30	-11	6	4	71	72	-2	8	4	66	-63	-6	11	4	33	-29
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-3	7	11	39	-33	-4	11	11	29	20	4	1	12	41	-37	-2	7	12	67	-62	8	2	13	37	47
-1	7	11	42	35	-3	11	11	37	-31	5	1	12	44	-42	1	7	12	31	-27	-8	3	13	33	37
0	7	11	58	-48	2	11	11	25	25	7	1	12	28	-34	2	7	12	45	-37	-7	3	13	33	-35

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
-6	3	13	31	33	2	8	13	61	53	-7	5	14	63	68	0	2	15	45	53	-4	1	16	42	-51
-5	3	13	33	38	5	8	13	58	55	-5	5	14	48	56	2	2	15	48	49	-3	1	16	36	-49
-2	3	13	56	58	8	8	13	30	28	-4	5	14	38	41	3	2	15	40	40	-1	1	16	33	-42
-1	3	13	37	-38	-5	9	13	42	43	-2	5	14	31	26	5	2	15	29	38	0	1	16	41	-48
1	3	13	38	39	-2	9	13	47	39	-1	5	14	37	40	0	3	15	43	-46	3	1	16	32	-39
2	3	13	29	-24	1	9	13	58	46	0	5	14	33	32	3	3	15	36	-43	-10	5	16	30	46
3	3	13	27	-23	-5	10	13	34	-28	2	5	14	71	68	-10	4	15	31	-50	-7	5	16	34	54
4	3	13	33	29	-4	10	13	35	-31	5	5	14	56	60	-7	4	15	48	-65	-4	5	16	46	55
6	3	13	34	-26	-1	10	13	60	-48	8	5	14	31	33	-4	4	15	62	-73	-3	5	16	35	42
-8	4	13	52	-59	2	10	13	43	-34	-5	6	14	44	44	-1	4	15	31	-33	-1	5	16	39	47
-7	4	13	31	-38	1	11	13	37	-29	-2	6	14	33	34	0	4	15	29	-29	3	5	16	44	46
-5	4	13	28	-27	-8	0	14	50	68	0	6	14	45	-48	2	4	15	54	-54	-3	6	16	33	-34
-4	4	13	54	-54	-5	0	14	56	69	1	6	14	36	39	3	4	15	30	-42	-4	7	16	36	-36
-2	4	13	45	-38	-2	0	14	57	58	-7	7	14	42	-42	5	4	15	29	-32	3	7	16	32	-35
-1	4	13	74	-73	0	0	14	64	-68	-4	7	14	51	-54	-7	8	15	44	52	-6	2	17	34	55
2	4	13	66	-64	3	0	14	34	-34	-1	7	14	60	-61	-4	8	15	63	67	-4	2	17	34	40
5	4	13	64	-68	-10	1	14	37	-57	2	7	14	42	-40	-1	8	15	33	37	-3	2	17	38	55
8	4	13	33	-34	-8	1	14	34	-46	-2	8	14	33	-30	0	8	15	39	34	0	2	17	40	47
-3	5	13	31	27	-7	1	14	69	-86	1	8	14	45	-42	2	8	15	45	45	-6	4	17	32	-48
-2	5	13	29	-27	-5	1	14	40	-48	-5	9	14	34	-30	3	8	15	35	39	-3	4	17	36	-45
1	5	13	44	-43	-4	1	14	60	-69	0	9	14	29	-24	5	8	15	33	26	0	4	17	33	-39
4	5	13	29	-30	-2	1	14	35	-32	-1	10	14	31	8	-6	9	15	30	-30	3	4	17	34	-34
-7	6	13	31	-26	-1	1	14	49	-52	-7	11	14	39	41	-3	9	15	38	-34	-3	8	17	32	36
-6	6	13	29	-24	2	1	14	64	-67	-5	11	14	34	33	-4	10	15	49	-44	0	8	17	35	36
3	6	13	27	-25	5	1	14	39	-44	-4	11	14	58	54	-1	10	15	31	-32	-5	0	18	30	-47
-2	7	13	28	-19	-8	2	14	31	-40	-1	11	14	43	40	-9	0	16	36	-55	1	0	18	28	-34
-8	8	13	47	50	-5	2	14	31	-31	2	11	14	41	34	-6	0	16	32	-46	-6	1	18	35	-59
-7	8	13	55	59	0	2	14	34	39	-10	2	15	44	69	-3	0	16	37	-36	-3	1	18	37	-53
-5	8	13	56	48	-2	3	14	30	-29	-7	2	15	55	79	0	0	16	35	-36	0	1	18	33	-41
-4	8	13	68	66	-5	4	14	30	-34	-4	2	15	58	67	-7	1	16	44	-62	0	5	18	37	48
-2	8	13	57	46	0	4	14	32	36	-1	2	15	50	59	-6	1	16	32	-47	-2	6	18	31	-32
-1	8	13	57	49	-10	5	14	35	45															

TABLE 4

Anisotropic thermal parameters ($/\mathring{A}^2 \times 10^3$)

Atom	$\underline{U}_{11}$	$\underline{U}_{22}$	$\underline{U}_{33}$	$\underline{U}_{23}$	$\underline{U}_{13}$	$\underline{U}_{12}$	$\underline{U}_{iso}$
F(1)	407(45)	246(32)	75(14)	-53(18)	100(21)	11(31)	155
F(2)	110(17)	174(22)	251(30)	53(21)	79(18)	-27(16)	144
F(3)	280(34)	201(26)	123(18)	105(19)	-3(20)	-48(23)	153
F(4)	126(19)	359(43)	239(30)	-57(30)	124(21)	15(24)	167
B	58(21)	97(24)	65(21)	2(18)	59(17)	0(17)	30
As(1)	40(1)	61(1)	27(1)	1(1)	14(1)	-2(1)	38
O(1)	51(10)	59(10)	44(9)	3(7)	11(8)	25(8)	46
C(111)	47(13)	41(12)	24(10)	-12(9)	16(9)	10(10)	28
C(112)	72(17)	64(15)	25(10)	10(10)	34(11)	2(13)	33
C(113)	74(17)	90(21)	39(13)	6(15)	33(12)	-1(17)	54
C(114)	82(19)	83(21)	39(14)	-6(14)	32(14)	-22(16)	55
C(115)	89(20)	66(16)	42(14)	-4(12)	30(14)	-52(15)	44
C(116)	71(17)	93(21)	23(11)	1k(12)	26(12)	-15(15)	43
C(121)	37(13)	56(14)	46(12)	-33(12)	14(10)	-27(12)	33
C(122)	40(13)	76(17)	32(12)	-15(11)	9(10)	-17(12)	42
C(123)	109(25)	89(21)	28(14)	-18(13)	8(15)	11(18)	60
C(124)	81(21)	93(21)	40(14)	-21(14)	5(14)	-13(18)	64
C(125)	31(12)	69(17)	72(16)	-8(15)	2(11)	-26(13)	47
C(126)	52(18)	75(19)	86(19)	-30(16)	16(14)	-29(14)	61
C(131)	45(15)	71(16)	46(14)	-20(13)	16(12)	-17(13)	48
C(132)	51(18)	110(26)	95(23)	-4(20)	-20(17)	-36(18)	71
C(133)	68(22)	110(29)	119(29)	-14(23)	-41(20)	-29(21)	83
C(134)	112(27)	107(26)	73(22)	-28(20)	41(20)	-70(24)	74
C(135)	89(24)	61(18)	71(19)	3(15)	23(18)	-8(16)	70
C(136)	56(15)	68(18)	47(12)	19(13)	0(11)	1(15)	54
As(2)	35(1)	61(1)	28(1)	9(1)	15(1)	-2(1)	36
O(2)	54(10)	62(10)	40(9)	-7(7)	14(8)	-4(8)	49
C(211)	38(13)	64(15)	32(12)	17(11)	10(10)	15(12)	38
C(212)	48(16)	74(19)	62(16)	-27(14)	5(12)	-5(14)	57
C(213)	95(30)	86(25)	139(33)	-38(22)	17(25)	8(21)	99
C(214)	97(25)	103(26)	52(18)	27(17)	19(18)	39(22)	71
C(215)	67(19)	92(23)	71(19)	3(19)	-12(15)	9(21)	75
C(216)	47(17)	96(23)	72(20)	10(17)	-15(14)	6(16)	66
C(221)	25(11)	91(18)	43(12)	34(13)	6(9)	10(12)	40
C(222)	55(15)	101(21)	59(16)	41(15)	31(13)	18(15)	55
C(223)	127(28)	91(21)	26(13)	0(13)	33(17)	14(20)	57
C(224)	106(27)	186(37)	44(17)	80(21)	1(17)	-18(25)	55
C(225)	31(16)	339(60)	72(20)	102(30)	4(15)	4(25)	77
C(226)	55(19)	234(43)	77(21)	87(25)	30(16)	17(24)	75
C(231)	38(13)	65(15)	26(13)	9(11)	15(10)	8(11)	36
C(232)	97(20)	48(14)	40(12)	3(10)	50(13)	-6(13)	40
C(233)/							

TABLE 4 (continued)

Atom	$\underline{U}_{11}$	$\underline{U}_{22}$	$\underline{U}_{33}$	$\underline{U}_{23}$	$\underline{U}_{13}$	$\underline{U}_{12}$	$\underline{U}_{iso}$
C(233)	102(20)	62(17)	35(12)	-10(13)	51(13)	2(16)	38
C(234)	56(17)	90(21)	52(16)	-1(15)	13(14)	15(16)	62
C(235)	119(26)	71(19)	110(24)	0(17)	88(22)	18(18)	70
C(236)	103(24)	82(21)	72(18)	18(15)	67(18)	32(17)	59

The anisotropic temperature factor takes the form

$$\exp[-2\pi^2(h^2\underline{a}^{*2}\underline{U}_{11}+k^2\underline{b}^{*2}\underline{U}_{22}+l^2\underline{c}^{*2}\underline{U}_{33}+2kl\underline{b}^{*}\underline{c}^{*}\underline{U}_{23}+2lh\underline{c}^{*}\underline{a}^{*}\underline{U}_{31}+2hk\underline{a}^{*}\underline{b}^{*}\underline{U}_{12})]$$

All phenyl hydrogens were allocated a common isotropic temperature factor which refined to 138(30) x 10^{-3}Å^2.

The column $\underline{U}_{iso}$ gives the geometric mean of the diagonal elements of the diagonalised matrix of $\underline{U}_{ij}$.

1,1,1,3,3,3-Hexaphenyldisilazane.

(i) Structure Factors.

(ii) Thermal Parameters.

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
4	0	0	64	65	9	2	0	47	47	6	4	0	9	8	8	6	0	14	11	12	8	0	19	19
6	0	0	70	66	10	2	0	20	17	9	4	0	8	5	10	6	0	5	3	13	8	0	15	12
8	0	0	65	62	11	2	0	16	19	10	4	0	8	9	12	6	0	34	27	15	8	0	11	21
10	0	0	68	61	12	2	0	9	7	11	4	0	8	8	13	6	0	12	11	16	8	0	8	10
12	0	0	11	15	13	2	0	12	11	12	4	0	33	33	14	6	0	50	48	18	8	0	8	13
14	0	0	25	29	15	2	0	34	31	13	4	0	22	22	15	6	0	27	28	19	8	0	15	18
16	0	0	44	39	16	2	0	14	10	14	4	0	51	48	16	6	0	17	13	21	8	0	9	15
18	0	0	19	18	17	2	0	28	26	15	4	0	23	21	17	6	0	27	27	2	9	0	31	35
20	0	0	29	31	18	2	0	10	12	16	4	0	16	13	18	6	0	13	13	3	9	0	28	30
22	0	0	14	14	19	2	0	19	19	19	4	0	13	12	20	6	0	11	13	4	9	0	18	18
2	1	0	95	103	20	2	0	13	13	21	4	0	13	15	21	6	0	8	10	5	9	0	14	16
3	1	0	49	52	21	2	0	22	25	22	4	0	19	20	22	6	0	11	15	6	9	0	10	5
4	1	0	4	5	22	2	0	16	15	23	4	0	13	12	1	7	0	23	21	7	9	0	34	29
5	1	0	69	70	23	2	0	13	14	2	5	0	18	21	2	7	0	16	14	9	9	0	14	11
6	1	0	30	26	1	3	0	69	64	3	5	0	28	23	3	7	0	20	24	12	9	0	19	21
7	1	0	5	2	2	3	0	31	26	4	5	0	55	57	5	7	0	31	34	15	9	0	11	14
8	1	0	26	24	3	3	0	82	82	5	5	0	6	10	6	7	0	7	2	17	9	0	19	16
9	1	0	36	38	4	3	0	31	28	6	5	0	10	8	7	7	0	10	9	18	9	0	12	15
10	1	0	28	27	5	3	0	90	89	8	5	0	12	8	10	7	0	7	7	19	9	0	8	11
11	1	0	24	27	6	3	0	46	38	9	5	0	14	9	13	7	0	37	31	20	9	0	12	12
13	1	0	9	12	7	3	0	20	20	10	5	0	14	16	14	7	0	11	9	0	10	0	71	58
15	1	0	8	6	8	3	0	9	12	11	5	0	9	5	16	7	0	8	10	2	10	0	28	26
16	1	0	19	19	9	3	0	26	24	12	5	0	35	41	17	7	0	25	23	3	10	0	19	17
17	1	0	22	18	10	3	0	9	7	13	5	0	10	8	19	7	0	15	17	4	10	0	27	26
18	1	0	30	31	11	3	0	16	15	15	5	0	18	16	0	8	0	27	27	5	10	0	16	13
19	1	0	19	22	12	3	0	6	8	16	5	0	24	26	1	8	0	31	35	6	10	0	13	5
21	1	0	8	8	13	3	0	8	11	18	5	0	10	11	2	8	0	7	4	8	10	0	11	9
0	2	0	46	47	16	3	0	25	22	20	5	0	19	22	3	8	0	12	13	9	10	0	14	19
1	2	0	97	105	17	3	0	33	36	0	6	0	34	31	4	8	0	34	38	10	10	0	9	6
2	2	0	33	32	19	3	0	28	33	1	6	0	9	10	5	8	0	15	18	11	10	0	20	21
3	2	0	143	147	21	3	0	12	13	2	6	0	43	40	6	8	0	23	25	13	10	0	15	16
4	2	0	78	73	0	4	0	31	27	3	6	0	16	15	7	8	0	8	10	14	10	0	9	7
5	2	0	38	34	1	4	0	32	32	4	6	0	20	16	8	8	0	8	10	16	10	0	25	27
6	2	0	17	15	3	4	0	17	16	5	6	0	53	48	9	8	0	17	13	20	10	0	11	14
7	2	0	71	63	4	4	0	24	23	6	6	0	33	32	10	8	0	19	22	1	11	0	23	21
8	2	0	36	35	5	4	0	11	6	7	6	0	26	24	11	8	0	23	16	2	11	0	44	41

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
8	11	0	8	5	5	0	1	36	35	2	2	1	80	76	23	3	1	14	16	1	6	1	20	19
9	11	0	9	6	6	0	1	46	44	3	2	1	47	48	0	4	1	54	47	2	6	1	54	55
11	11	0	15	15	7	0	1	4	4	4	2	1	32	31	1	4	1	57	53	3	6	1	22	19
12	11	0	8	12	8	0	1	28	29	5	2	1	13	10	2	4	1	23	23	4	6	1	48	43
13	11	0	10	11	9	0	1	11	10	6	2	1	24	26	3	4	1	45	45	5	6	1	58	54
17	11	0	8	10	10	0	1	8	9	8	2	1	24	24	4	4	1	52	44	6	6	1	54	49
18	11	0	11	14	11	0	1	25	23	9	2	1	25	25	5	4	1	62	57	7	6	1	27	27
19	11	0	11	12	12	0	1	15	16	10	2	1	47	43	6	4	1	31	32	8	6	1	22	23
0	12	0	26	24	14	0	1	19	20	11	2	1	12	10	7	4	1	43	41	9	6	1	15	16
2	12	0	19	19	15	0	1	20	16	12	2	1	7	7	8	4	1	25	25	10	6	1	14	19
5	12	0	16	15	16	0	1	14	17	13	2	1	17	18	9	4	1	62	55	11	6	1	28	33
6	12	0	11	10	17	0	1	30	29	14	2	1	19	22	10	4	1	22	22	12	6	1	19	17
7	12	0	18	16	18	0	1	18	20	16	2	1	29	26	11	4	1	22	27	13	6	1	17	13
9	12	0	31	27	19	0	1	14	15	17	2	1	21	21	12	4	1	20	21	14	6	1	32	28
11	12	0	11	9	2	1	1	77	85	18	2	1	12	10	15	4	1	37	32	15	6	1	9	6
12	12	0	19	20	3	1	1	63	66	20	2	1	10	11	18	4	1	12	9	16	6	1	12	12
14	12	0	11	11	4	1	1	63	62	21	2	1	10	8	20	4	1	16	15	17	6	1	9	6
15	12	0	14	16	5	1	1	13	11	0	3	1	134	140	0	5	1	49	47	18	6	1	16	17
17	12	0	8	11	6	1	1	30	27	1	3	1	114	116	1	5	1	34	31	22	6	1	12	13
3	13	0	19	19	7	1	1	21	19	2	3	1	107	105	2	5	1	51	49	1	7	1	46	45
5	13	0	23	20	8	1	1	64	62	3	3	1	98	96	3	5	1	109	106	2	7	1	17	13
6	13	0	12	10	9	1	1	8	7	4	3	1	8	9	4	5	1	65	62	3	7	1	20	22
8	13	0	13	14	10	1	1	42	39	5	3	1	12	15	5	5	1	45	41	4	7	1	23	25
10	13	0	11	10	11	1	1	25	24	6	3	1	31	27	6	5	1	25	21	5	7	1	28	25
11	13	0	8	11	13	1	1	12	12	7	3	1	42	44	7	5	1	69	59	6	7	1	18	18
1	14	0	15	15	14	1	1	10	11	8	3	1	29	31	8	5	1	29	29	7	7	1	5	5
3	14	0	16	20	15	1	1	19	17	9	3	1	42	40	9	5	1	18	20	8	7	1	30	33
13	14	0	9	14	16	1	1	15	13	10	3	1	28	28	10	5	1	27	27	9	7	1	24	28
14	14	0	9	11	17	1	1	14	16	11	3	1	37	39	12	5	1	32	37	10	7	1	26	27
2	15	0	20	19	18	1	1	22	21	12	3	1	32	35	13	5	1	38	31	11	7	1	36	32
4	15	0	12	16	19	1	1	13	14	13	3	1	19	24	14	5	1	26	24	12	7	1	54	47
7	15	0	8	12	20	1	1	14	15	14	3	1	16	12	15	5	1	17	14	13	7	1	22	22
9	15	0	10	13	22	1	1	13	15	15	3	1	33	26	17	5	1	13	12	14	7	1	26	24
2	0	1	31	31	23	1	1	10	10	16	3	1	14	13	19	5	1	9	11	15	7	1	9	10
3	0	1	9	10	0	2	1	44	44	17	3	1	16	16	20	5	1	9	10	16	7	1	9	7
4	0	1	115	128	1	2	1	44	41	21	3	1	15	16	21	5	1	8	8	17	7	1	10	8

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
18	7	1	13	17	1	10	1	31	24	16	12	1	8	11	17	0	2	12	11	17	2	2	28	26
20	7	1	12	12	2	10	1	9	8	19	12	1	9	8	18	0	2	13	11	19	2	2	17	18
0	8	1	22	20	3	10	1	39	32	1	13	1	9	10	1	1	2	85	84	20	2	2	10	9
1	8	1	16	17	4	10	1	21	19	2	13	1	13	14	2	1	2	139	147	23	2	2	9	8
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5	6	8	21	23	4	9	8	7	6	8	0	9	16	18	10	3	9	12	16	6	6	9	20	20
6	6	8	9	8	5	9	8	12	19	9	0	9	15	12	12	3	9	9	10	8	6	9	13	14
7	6	8	13	12	7	9	8	9	8	10	0	9	10	10	13	3	9	8	9	9	6	9	13	14
10	6	8	10	9	8	9	8	18	16	11	0	9	17	15	14	3	9	10	7	10	6	9	11	11
14	6	8	14	12	9	9	8	10	9	12	0	9	8	8	2	4	9	6	4	11	6	9	13	14
15	6	8	11	8	10	9	8	10	10	15	0	9	8	10	3	4	9	13	15	12	6	9	7	7
16	6	8	10	10	11	9	8	12	10	17	0	9	11	9	4	4	9	9	12	13	6	9	14	12
17	6	8	8	8	12	9	8	13	13	4	1	9	6	7	5	4	9	25	25	14	6	9	10	10
20	6	8	8	8	14	9	8	10	11	5	1	9	6	4	6	4	9	20	21	1	7	9	7	6
0	7	8	9	10	0	10	8	20	20	6	1	9	8	14	7	4	9	13	14	2	7	9	9	12

H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC	H	K	L	FO	FC
3	7	9	9	10	2	11	9	11	10	14	3	10	9	10	7	7	10	16	20	8	1	11	5	6
4	7	9	8	8	7	11	9	11	9	3	4	10	6	5	8	7	10	7	8	9	1	11	5	4
5	7	9	10	10	8	11	9	11	10	5	4	10	14	16	9	7	10	9	10	10	1	11	5	6
6	7	9	22	25	9	11	9	11	9	6	4	10	11	15	13	7	10	13	9	11	1	11	6	6
7	7	9	7	8	4	12	9	13	12	7	4	10	8	11	1	8	10	9	8	13	1	11	7	9
8	7	9	13	12	6	12	9	11	10	9	4	10	17	21	3	8	10	7	7	14	1	11	8	10
10	7	9	11	12	10	12	9	12	13	10	4	10	18	19	4	8	10	15	16	16	1	11	9	6
11	7	9	9	9	6	13	9	9	8	11	4	10	6	6	5	8	10	14	15	17	1	11	12	9
12	7	9	13	11	8	13	9	10	10	12	4	10	11	13	9	8	10	7	8	8	2	11	6	6
13	7	9	10	10	6	0	10	8	5	16	4	10	9	8	11	8	10	11	7	10	2	11	6	10
14	7	9	13	11	7	0	10	4	5	18	4	10	11	8	12	8	10	8	6	11	2	11	8	11
1	8	9	12	11	11	0	10	11	12	19	4	10	13	10	2	9	10	9	6	14	2	11	9	10
2	8	9	9	9	12	0	10	10	11	2	5	10	8	9	3	9	10	8	8	15	2	11	7	8
4	8	9	14	17	17	0	10	8	8	4	5	10	5	7	5	9	10	8	8	16	2	11	12	9
5	8	9	7	7	5	1	10	5	5	6	5	10	13	15	7	9	10	14	17	5	3	11	5	4
6	8	9	7	8	7	1	10	20	24	8	5	10	14	13	8	9	10	12	10	6	3	11	5	8
7	8	9	18	19	8	1	10	15	14	9	5	10	6	4	3	10	10	6	6	9	3	11	6	4
9	8	9	10	10	9	1	10	10	8	10	5	10	8	9	4	10	10	12	10	10	3	11	6	5
10	8	9	21	18	10	1	10	9	13	12	5	10	8	9	5	10	10	16	12	12	3	11	6	5
11	8	9	12	10	11	1	10	8	13	14	5	10	14	12	9	10	10	9	8	18	3	11	12	11
12	8	9	15	13	12	1	10	9	10	0	6	10	5	2	10	10	10	13	12	7	4	11	6	8
2	9	9	10	11	13	1	10	9	10	2	6	10	7	7	12	10	10	9	7	15	4	11	10	6
5	9	9	6	7	14	1	10	12	13	3	6	10	12	12	1	11	10	9	8	4	5	11	7	7
6	9	9	8	7	15	1	10	6	6	4	6	10	5	5	8	11	10	9	8	5	5	11	8	5
7	9	9	15	19	7	2	10	5	9	5	6	10	11	10	2	12	10	10	8	10	5	11	6	6
9	9	9	15	13	8	2	10	5	8	6	6	10	12	11	9	12	10	10	10	0	6	11	10	9
13	9	9	14	11	9	2	10	11	15	8	6	10	8	8	0	13	10	11	10	2	6	11	7	7
15	9	9	11	8	10	2	10	11	12	9	6	10	6	6	7	13	10	9	9	3	6	11	8	11
2	10	9	7	7	11	2	10	13	11	10	6	10	9	11	4	14	10	9	9	4	6	11	5	9
3	10	9	8	6	7	3	10	20	24	11	6	10	9	8	6	0	11	5	8	5	6	11	6	5
4	10	9	12	8	8	3	10	9	9	12	6	10	8	10	9	0	11	7	10	7	6	11	7	5
5	10	9	12	11	9	3	10	8	11	13	6	10	9	8	12	0	11	8	9	1	7	11	7	7
6	10	9	10	7	10	3	10	7	6	17	6	10	10	9	14	0	11	6	6	2	7	11	16	17
8	10	9	16	16	11	3	10	8	8	0	7	10	11	8	15	0	11	11	13	4	7	11	11	11
9	10	9	15	14	12	3	10	7	7	2	7	10	15	15	16	0	11	10	9	5	7	11	8	11
11	10	9	13	13	13	3	10	10	11	3	7	10	8	9	7	1	11	7	12	6	7	11	8	8

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Table A. Anisotropic temperature parameters ($\mathring{A}^2 \times 10^3$)

Atom	U_{11}	U_{22}	U_{33}	U_{23}	U_{31}	U_{12}
Si(1)	32(1)	31(1)	38(1)	5(1)	3(1)	4(1)
Si(2)	32(1)	29(1)	40(1)	-1(1)	-2(1)	2(1)
N	59(5)	43(4)	46(5)	-10(3)	-14(4)	26(3)
C(11)	35(4)	30(4)	42(5)	3(4)	2(4)	12(3)
C(12)	58(6)	29(4)	86(8)	13(5)	-6(5)	5(5)
C(13)	74(7)	23(5)	156(14)	1(7)	-28(9)	-6(5)
C(14)	67(7)	58(7)	67(8)	-7(6)	8(6)	-23(6)
C(15)	57(6)	54(6)	68(8)	4(5)	-7(5)	-1(5)
C(16)	45(5)	36(5)	62(8)	-7(5)	-11(5)	1(4)
C(21)	35(5)	36(4)	57(6)	-1(4)	17(4)	13(4)
C(22)	55(7)	113(10)	72(9)	-36(7)	12(6)	-36(7)
C(23)	78(9)	194(18)	102(13)	-76(13)	45(9)	-83(10)
C(24)	136(14)	87(10)	166(18)	-55(11)	99(14)	-70(10)
C(25)	127(12)	82(9)	73(11)	14(7)	33(9)	-5(9)
C(26)	67(7)	74(7)	59(8)	21(6)	17(6)	9(5)
C(31)	47(5)	34(4)	39(6)	4(4)	-4(4)	6(4)
C(32)	44(5)	44(6)	108(10)	2(6)	29(6)	14(4)
C(33)	59(7)	71(8)	135(13)	5(9)	37(8)	21(6)
C(34)	64(7)	53(6)	82(9)	-5(6)	-5(6)	39(5)
C(35)	120(10)	32(6)	122(12)	24(7)	21(10)	40(6)
C(36)	81(7)	33(5)	119(11)	20(6)	40(8)	17(5)
C(41)	35(4)	26(4)	45(5)	1(3)	2(4)	3(3)
C(42)	44(5)	33(5)	85(8)	3(5)	-13(5)	-8(4)
C(43)	55(6)	38(6)	103(10)	6(6)	3(6)	-3(5)
C(44)	63(6)	29(4)	102(9)	12(6)	34(7)	2(5)
C(45)	85(8)	49(6)	69(8)	17(5)	19(6)	8(6)
C(46)	48(5)	53(6)	58(7)	13(5)	-7(4)	-3(4)
C(51)	32(4)	41(5)	56(6)	3(4)	-7(4)	2(4)
C(52)	62(6)	45(5)	70(8)	-10(5)	13(5)	2(5)
C(53)	81(8)	69(8)	120(12)	-27(8)	30(9)	27(7)
C(54)	87(9)	47(6)	91(10)	-19(6)	-28(7)	20(6)
C(55)	80(7)	25(4)	136(11)	8(7)	-2(9)	-20(5)
C(56)	72(6)	31(4)	78(8)	-5(5)	0(6)	2(4)
C(61)	37(4)	29(4)	57(6)	1(4)	-4(4)	1(4)
C(62)	50(6)	56(6)	89(9)	-27(6)	-19(6)	19(5)
C(63)	40(6)	102(9)	108(11)	-39(8)	2(6)	39(6)
C(64)	33(5)	79(7)	67(8)	9(6)	-3(5)	10(5)
C(65)	46(6)	49(6)	93(9)	-11(6)	-24(6)	-4(5)
C(66)	42(5)	49(5)	55(7)	-14(5)	-12(4)	6(4)

The anisotropic temperature factor takes the form:

$$\exp[-2\pi^2(h^2\underline{a}^{*2}\underline{U}_{11} + k^2\underline{b}^{*2}\underline{U}_{22} + l^2\underline{c}^{*2}U_{33} + 2kl\underline{b}^{*}\underline{c}^{*}\underline{U}_{23} + 2lh\underline{c}^{*}a^{*}\underline{U}_{31} + 2hka^{*}b^{*}\underline{U}_{12})]$$

The phenyl hydrogens had a common isotropic temperature factor:

$73(8) \times 10^{-302}$

The unique H(N) hydrogen had an isotropic temperature factor:

$14(4) \times 10^{-202}$